

The Huckleberry Ridge Tuff, Yellowstone: evacuation of multiple magmatic systems in a complex episodic eruption

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SAMPLING AND ANALYTICAL METHODS

Sampling methodology

Previous work on the Huckleberry Ridge Tuff (HRT) has almost exclusively used whole-rock samples that have the limitations in that they can only be associated with an eruptive unit and not with any specific magma composition. The focus for this study was placed on sampling single clasts, and particularly glassy juvenile clasts where possible. Due to the dominantly welded and devitrified nature of the HRT, occurrence of glassy material and non-welded material is severely limited. Locations where these occurred (Fig. 1; Electronic Appendix 1) were intensively sampled. Although not representative of the full spatial extent of the HRT deposits, a suite of samples representative of the range of the physical nature of the clasts were collected where possible (see main paper and Table 1 there for discussion of clast types). This was repeated for members A, B and C of the ignimbrite to enable investigation of any inter-member variation of physically similar clasts. Where clasts were sampled from welded ignimbrite, only partial removal was often possible, although enough of the clast to be representative of the overall physical characteristics was sampled.

The initial fall deposits were sampled at Mt. Everts (locality 5: Electronic Appendix 1), and sampling details and results are published in Myers *et al.* (2016) and Swallow *et al.* (2018).

General sample preparation

Single juvenile clasts (glassy pumice for preference, where available) were sampled in order to determine the nature of the erupted material and to be able to discern any heterogeneities present. Samples were trimmed and washed to remove any adhering matrix, then systematically treated to ensure enough material was available for the variety of techniques used in this study (Fig. A).

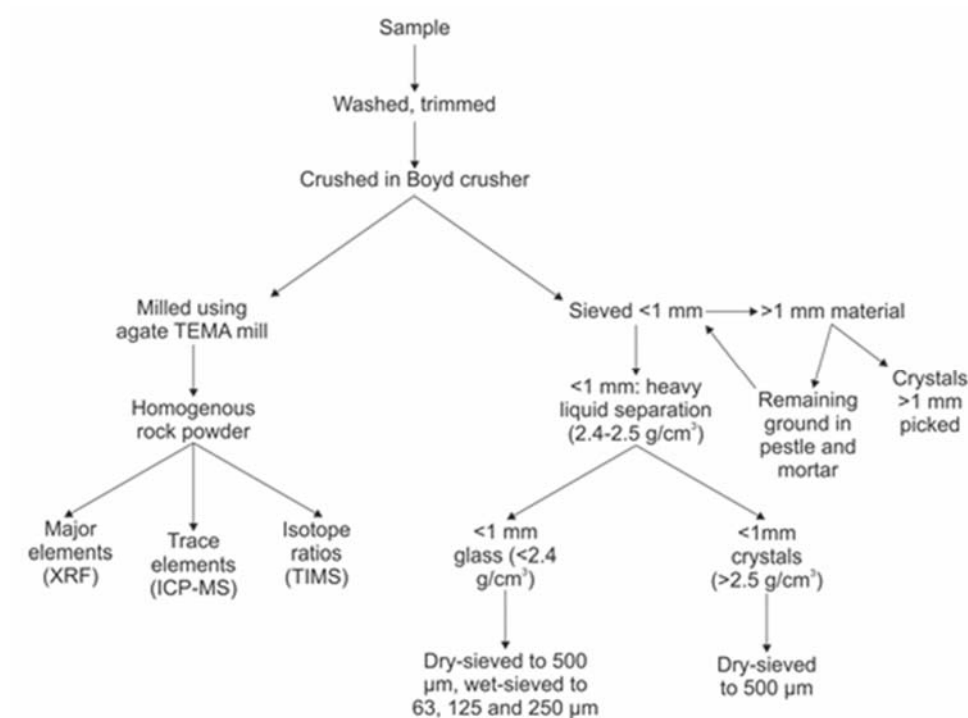


Fig. A: Flow chart showing general sample preparation for the variety of techniques used in this study

Cleaned samples were coarsely crushed in a Rocklabs Boyd crusher to ~2 mm or the size of the largest crystals, whichever was larger. The advantage of using a Boyd crusher is that it liberates crystals from pumices intact, rather than fracturing them as often occurs with impact crushing. The crushed sample was then manually split into two parts, one for bulk analysis representative of the single-clast and the other to be further processed for component-specific analysis. The single clast portion was then ground to a fine powder using an agate TEMA ring mill. The remaining sample was sieved to 1 mm. Coarser material was gently ground by hand using a ceramic mortar and pestle to liberate any remaining crystals intact and to reduce the size of any matrix to <1 mm so the sample could be separated using heavy-liquid techniques. Crystals >1 mm were hand-picked. Crystals (>2.5 g/cm³) and rhyolite glass (<2.4 g/cm³) were separated from the sieved fraction using lithium-polytungstate (LST) heavy-liquid with a reduced density of 2.4-2.5 g/cm³. The crystal fraction was weighed to give an approximate wt% crystals for each sample. Crystals were sieved again to give a 0.5-1 mm fraction, which was found to be sufficiently large to encompass any textural variety observed in the crystal cargo. The glass portion was dry- and wet-sieved into 63, 125, 250 and 500 µm

size fractions, and glass shards in the 250-500 μm fraction were hand-picked for *in situ* micro-analysis.

SINGLE-CLAST COMPOSITIONS

XRF: single-clast major and minor elements

Single clast major and selected trace-element analyses were conducted at the Open University, United Kingdom (OU), the University of Auckland, New Zealand (UA), and the University of Waikato (UW). Any samples yielding loss on ignition (LOI) values >6 wt% were removed from the dataset as they were suspected/confirmed to contain carbonate contaminant. XRF data were normalised to 100% volatile-free. Replicate analysis of multiple standards in each laboratory return individual 2 s.d. relative precisions of better than 3% for almost all major element oxides, with most better than 1%. Offsets from preferred or recommended values are generally $<5\%$ for all elements. Analytical 2 s.d. precisions for Ba are mostly better than 6% with offsets $<7\%$ from preferred or recommended values (Table S1). Duplicates analysed between laboratories show offsets $<5\%$ for all element oxides >0.1 wt% and $\leq 5\%$ for Ba. Complete standards data are given in Electronic Appendix 3.

Solution ICP-MS: trace elements

Selected single clast samples, from the same powders that were used for XRF analysis, were prepared for trace element analysis by inductively coupled plasma mass spectrometry (ICP-MS) at Victoria University of Wellington (VUW), New Zealand. All sample preparation took place in the ultra-clean geochemistry facility, with air filtered to Class 100 or better and only ultra-purity acids (key metals <10 ppt) were used in sample preparation. Acids used in the initial stages of beaker cleaning were analytical reagent (AR) grade. Acids were diluted with Milli-Q water (MQ) and molarities checked with a density meter. Due to the presence of zircon in many samples in this study, and the inefficient breakdown of this refractory mineral by standard low-pressure digestion techniques (Hu *et al.*, 2013, and references therein), two methods were used in sample preparation for solution ICP-MS analysis.

Hydrofluoric acid digestion

Samples with $<\sim 65$ wt% SiO_2 and inferred to contain no or minimal zircon were prepared using standard HF-HNO₃ techniques. Approximately 50 mg of sample powder was weighed

into pre-cleaned 23 ml Savillex screw-top Teflon beakers using a high precision balance (to ± 0.0001 g). Subsequently, ~ 2 ml concentrated hydrofluoric acid (29 M HF) and ~ 0.5 ml concentrated nitric acid (16 M HNO_3) were added to the powders. The beakers were then capped and placed on a hotplate at 120°C for 3-4 days to ensure complete digestion of the samples. The samples were visually inspected to check that no solid materials remained. Following digestion, the solution was evaporated to incipient dryness to prevent sample loss and formation of insoluble fluorides. The sample cake was then refluxed twice in ~ 2 ml concentrated HNO_3 and taken to incipient dryness. Subsequently ~ 5 ml 6M hydrochloric acid (HCl) was added and the beakers placed on a hotplate overnight. Following complete dissolution, the HCl was evaporated to dryness and the sample taken up in ~ 2 ml concentrated HNO_3 for 2-3 days. After further evaporation, the sample was refluxed in 9 ml 1M HNO_3 for 2-3 days. The final solutions were run through a centrifuge at 2000 rpm for 5 minutes to ensure no solids remained before being transferred into pre-cleaned 10 ml polyethylene centrifuge-tubes (c-tube) and precisely weighed (± 0.0001 g), ensuring all of the solution was washed from the beaker. The sample was further diluted by taking a 50 μl aliquot of the final solution and transferring into another pre-cleaned polyethylene c-tube and adding ~ 9 ml 1% HNO_3 . The c-tube was weighed prior to and following the addition of all solutions to enable the calculation of the final dilution factor, usually $\sim 32,000$ -fold. International rock standards (BHVO-2 and BCR-2) and a procedural blank were processed in the same way along with each sample batch.

Ammonium fluoride digestion

Following the procedure of Hu *et al.* (2013), approximately 50 mg of powder was weighed in pre-cleaned 60 ml Savillex screw top Teflon beakers. High-purity ammonium fluoride (NH_4F ; Alfa-Aesar) was added to yield a sample: NH_4F ratio of 1:6. A few drops of MQ water were added to each sample and the beakers were shaken to ensure all the powder was wetted. The beakers were then placed in a Thermo Scientific Heratherm oven at 210°C for 60 hours, a temperature and duration chosen to avoid softening of the Teflon at higher temperatures, yet ensuring complete digestion of the powder.

Table S1: Accuracy and precision of repeated analysis of standard materials and samples by XRF.												
		SiO₂	TiO₂	Al₂O₃	Fe₂O₃(T)	MnO	MgO	CaO	Na₂O	K₂O	P₂O₅	Ba
WS-E n=16 Jochum <i>et al.</i> (2016)	Preferred	51.07	2.43	13.77	13.09	0.17	5.63	9.00	2.48	1.00	0.30	335.00
	Average	51.11	2.42	13.83	13.14	0.17	5.55	8.98	2.43	1.00	0.30	330.79
	% Offset	0.08	-0.13	0.46	0.36	1.39	-1.35	-0.17	-2.08	-0.03	-1.01	-1.26
	2 s.d.	0.14	0.01	0.07	0.03	0.00	0.04	0.06	0.06	0.01	0.01	18.09
	2 s.d.%	0.27	0.61	0.49	0.25	1.79	0.66	0.67	2.66	1.44	2.44	5.47
OU-3 n=16 Long-term OU average	Preferred	74.09	0.22	11.11	3.83	0.09	-	0.20	3.68	4.55	-	29.00
	Average	74.15	0.22	11.03	3.87	0.09	0.03	0.21	3.72	4.57	0.02	30.04
	% Offset	0.09	0.36	-0.73	1.16	2.14		4.47	1.12	0.43		3.60
	2 s.d.	0.21	0.01	0.06	0.04	0.00	0.01	0.01	0.08	0.03	0.00	12.65
	2 s.d.%	0.29	2.36	0.52	0.92	2.93	38.83	3.47	2.24	0.65	14.62	42.12
SY-2 n=12 Gladney & Roelandts (1990)	Preferred	60.11	0.15	12.04	6.31	0.32	2.69	7.96	4.45	4.31	0.43	464.00
	Average	59.81	0.13	11.97	6.19	0.32	2.62	7.90	4.46	4.22	0.43	435.00
	% Offset	-0.51	-11.33	-0.60	-1.98	-1.48	-2.65	-0.75	0.30	-2.09	-0.33	-6.25
	2 s.d.	0.17	0.00	0.06	0.02	0.01	0.05	0.02	0.01	0.01	0.00	13.49
	2 s.d.%	0.28	2.38	0.53	0.25	2.97	2.05	0.21	0.33	0.34	1.09	3.10
JA-2 n=10 Jochum <i>et al.</i> (2016)	Preferred	56.39	0.67	15.51	6.29	0.11	7.84	6.26	1.78	3.07	0.15	308.40
	Average	56.46	0.67	15.64	6.11	0.11	7.21	6.15	1.80	3.10	0.15	314.20
	% Offset	0.12	-0.43	0.83	-2.86	-3.85	-8.10	-1.77	1.29	1.04	0.72	1.88
	2 s.d.	0.28	0.01	0.06	0.04	0.00	0.13	0.03	0.02	0.01	0.00	6.68
	2 s.d.%	0.50	1.06	0.36	0.59	4.26	1.84	0.54	0.97	0.32	1.31	2.13
Duplicates	YR253	50.40	1.92	16.49	12.65	0.17	5.66	8.72	2.87	0.81	0.29	406
	YR253D	50.52	1.97	16.37	12.66	0.17	5.64	8.67	2.89	0.82	0.28	427
	% Offset	0.23	2.27	-0.71	0.05	1.99	-0.44	-0.60	0.78	0.56	-1.34	5.20
	YR275	74.48	0.04	14.90	0.25	0.01	0.14	1.59	4.33	4.19	0.06	4181
	YR275D	74.67	0.03	14.96	0.24	0.00	0.14	1.55	4.14	4.21	0.05	4118
	% Offset	0.25	-14.88	0.44	-4.59	-45.84	-3.93	-2.36	-4.55	0.44	-13.31	-1.50
WS-E and OU-3 analysed at the Open University, UK and SY-2 and JA-2 at the University of Auckland, New Zealand. Duplicates run between both laboratories. Oxide abundances in wt%, Ba in ppm. Full standard data given in Electronic Appendix 3.												

Once the beakers had cooled, approximately 4 ml concentrated HNO₃ (or enough to cover the base of the beakers) was added and immediately dried down on a hot plate at 160 °C. This step was repeated before the samples were refluxed in 5 ml of 6 M HNO₃ overnight at 120 °C. The resulting limpid solution was transferred to a c-tube, weighed and diluted with 1 % HNO₃ to a 10,000-fold dilution for analysis. International standards GSP-2 and JR-1, and a procedural blank were processed alongside each batch of samples.

Analysis and correction

Trace element measurements were carried out using a Thermo-Fisher Element2 sector-field ICP-MS at VUW equipped with an ESI autosampler. Analysis procedure was similar to that of Eggins *et al.* (1997). The ICP-MS was tuned using a 1 ppb multi-element standard, and the torch position, gas flows and ion optics adjusted to optimise performance (i.e. low relative standard deviations of <1-2%; low oxide abundances: UO⁺/U⁺ <6-7 %, BaO⁺/Ba⁺ <2 % and Ba⁺⁺/Ba⁺ <5 %). Some isotopes were analysed at higher resolution to resolve the relevant peak (e.g. ⁴⁵Sc, ⁴⁷Ti, ⁵¹V) from neighbouring interferences, or to attenuate the count rates for highly abundant isotopes (e.g. ²⁵Mg, ⁴³Ca, ⁵⁵Mn). Solutions were analysed for 90 s and were preceded by a 120 s washout using 1 % HNO₃.

Concentrations of samples prepared using conventional HF digestion and published in Swallow *et al.* (2018b) were calculated by correcting to an identically prepared rock standard of known composition (BHVO-2). Unknown samples (blocks of 3) were bracketed by repeated analyses of BHVO-2. Repeated analyses of a secondary standard, BCR-2, used to determine accuracy of the measurements and to monitor drift, were done after each block of unknowns. A procedural blank was analysed at the beginning of each run, with counts recorded being within the range of background levels for all elements. Abundances of elements were calculated using the following calculations:

$$Sample\ C_i = (Sample_{CPS} / BHVO-2_{CPS}) \times (Sample_{Dil} / BHVO-2_{Dil}) \times ref. \quad (1)$$

$$Sample\ C_i\ (Ca\ corr.) = eq.\ (1) \times (C_{Ca}\ XRF / C_{Ca}\ ICP-MS) \quad (2)$$

where C_i and C_{Ca} are the concentrations of the measured isotope i and Ca, respectively, and X_{Dil} is the dilution factor of the relevant solution. Reference values for BHVO-2 and BCR-2 were taken from Jochum *et al.* (2016). Repeated analyses of BCR-2 was used to estimate external precisions, which were <6-7% for most elements except Li, Nb, Cs, Lu, Ta, Tl, Pb,

Th, U, Ni Cu, Zn (<20%) and Mo (>20%). Offsets relative to preferred values are <6-7% for all elements, apart from Ta, Tl, Cu (<15%) and Mo (>15%: Table S2; Electronic Appendix 3). Duplicate samples analysed within and between runs show 2 s.d.% precisions of typically <10 % for all elements except Zn, Nb and Th (<15 %: Table S3).

Table S2: Accuracy and precision of repeated analyses of BCR-2 by ICP-MS.							
	Preferred	Average	% Offset	2 s.d.	2 s.d.%	BCR-2 (NH₄F)	NH₄F % offset
Li	9.13	9.34	2.34	0.81	8.68	8.41	-7.92
Sc	33.53	33.34	-0.58	0.99	2.96	31.25	-6.79
V	417.60	416.93	-0.16	10.88	2.61	439.31	5.20
Cr	15.85	14.79	-6.66	0.55	3.75	16.24	2.46
Mn	0.20	0.20	0.81	0.00	1.85	0.17	-15.10
Co	37.33	37.72	1.05	0.73	1.93	40.32	8.02
Ni	12.57	11.82	-5.96	0.94	7.95	14.77	17.46
Cu	19.66	16.93	-13.87	1.44	8.48		
Zn	129.50	136.03	5.05	19.39	14.25		
Ga	22.07	21.96	-0.51	0.81	3.69	23.13	4.79
Rb	46.02	48.16	4.65	1.25	2.60	46.80	1.70
Sr	337.40	338.55	0.34	10.35	3.06	338.03	0.19
Y	36.07	35.77	-0.82	0.80	2.24	33.41	-7.39
Zr	186.50	188.39	1.02	4.95	2.63	197.27	5.78
Nb	12.44	12.95	4.08	2.09	16.17	12.26	-1.42
Mo	250.60	380.16	51.70	634.33	166.86		
Cs	1.16	1.19	2.28	0.10	8.36	1.19	2.30
Ba	683.90	693.57	1.41	31.43	4.53	688.79	0.72
La	25.08	24.95	-0.51	0.96	3.85	25.52	1.75
Ce	53.12	52.92	-0.39	2.46	4.64	54.59	2.77
Pr	6.83	6.81	-0.30	0.29	4.28	6.81	-0.19
Nd	28.26	28.31	0.17	1.39	4.92	28.36	0.37
Sm	6.55	6.50	-0.71	0.29	4.43	6.64	1.50
¹⁵¹ Eu	1.99	2.01	0.82	0.15	7.26	2.12	6.37
¹⁵³ Eu	1.99	2.08	4.34	0.15	6.99	2.15	8.08
Gd	6.81	6.81	-0.06	0.41	5.98	6.70	-1.68
Tb	1.08	1.05	-2.39	0.06	5.41	1.11	3.43
Dy	6.42	6.31	-1.76	0.38	6.01	6.34	-1.24
Ho	1.31	1.29	-2.06	0.08	5.93	1.27	-3.52
Er	3.67	3.55	-3.25	0.25	7.12	3.77	2.68
Tm	0.53	0.52	-2.40	0.03	6.65	0.51	-4.27
Yb	3.39	3.30	-2.66	0.23	6.95	3.32	-2.02
Lu	0.50	0.49	-3.15	0.04	7.59		
Hf	4.97	4.93	-0.77	0.23	4.75	5.26	5.75
Ta	0.79	0.85	8.36	0.08	9.86		
Tl	0.27	0.29	7.20	0.05	16.14		
Pb	10.59	10.93	3.19	2.04	18.65	9.77	-7.76
Th	5.83	5.64	-3.30	0.66	11.72	5.99	2.76
U	1.68	1.63	-3.44	0.13	8.26	1.63	-3.43
Average derived from 19 analyses using HF digestion techniques. Single measurement from a NH ₄ F digestion of BCR-2. Concentrations in ppm except Mn in wt%. Preferred values from Jochum <i>et al.</i> (2016). Full standard data given in Electronic Appendix 3.							

Table S3: Repeated samples analysed by ICP-MS using HF digestion techniques. Concentrations in ppm except Mn (wt%).																	
	YP188	YP188	YP188	2s.d.%	YP244	YP244	2s.d.%	YP334B	YP334B	YP334B	2s.d.%	YP382	YP382	2s.d.%	YR425	YR425	2s.d.%
Li	22.14	22.42	21.89	1.97	5.39	5.06	6.34	21.24	18.44	21.40	0.77	21.79	26.13	18.14	17.42	15.79	9.82
Sc	16.91	16.86	17.34	2.53	21.06	21.11	0.22	26.28	26.35	25.51	2.95	37.93	38.74	2.12	22.69	22.42	1.19
V	25.12	26.11	25.74	3.19	69.05	68.70	0.50	79.71	76.34	79.17	0.67	119.9	124.06	3.34	103.9	102.02	1.86
Cr	3.88	3.80	3.75	2.84	5.89	6.08	3.04	5.38	3.62	4.51	17.6	7.31	7.18	1.83	2.90	2.90	0.04
M	0.14	0.16	0.15	7.20	0.11	0.11	0.34	0.22	0.22	0.22	3.09	0.06	0.06	3.16	0.27	0.26	3.00
Co	13.38	14.99	14.76	9.88	11.30	11.16	1.25	20.58	19.75	20.86	1.39	12.27	12.55	2.26	31.19	30.14	3.42
Ni	2.41	2.31	1.99	16.16	5.98	5.84	2.40	5.17	5.43	5.20	0.54	3.99	3.63	9.49	3.33	3.02	9.86
Zn	174	199	210	15.31	231	255	10.06	194	190	217	11.2	330	349	5.45	181	196	7.95
Ga	26.75	27.22	27.83	3.24	26.01	26.02	0.06	24.26	24.09	25.21	3.86	28.79	29.70	3.12	24.34	23.95	1.59
Rb	98.62	102.05	99.65	2.87	62.92	61.46	2.36	68.00	67.36	67.95	0.07	46.99	47.93	1.98	33.75	33.18	1.69
Sr	138	138	139	0.33	247	247	0.05	265	267	272	2.42	338	344	1.84	347	334	3.82
Y	313	293	294	6.13	74	76	2.41	73	70	72	0.91	108	113	4.28	92	88	3.64
Zr	792	827	814	3.56	1386	1334	3.82	1353	1367	1407	3.92	1755	1760	0.25	753	744	1.20
Nb	44.37	48.55	46.02	7.44	42.57	36.67	14.89	38.98	33.27	36.75	5.89	35.44	39.23	10.16	61.45	64.63	5.05
Cs	1.58	1.68	1.70	6.15	0.76	0.75	1.55	0.98	0.95	1.04	6.41	0.67	0.73	9.71	0.52	0.49	5.80
Ba	2722	2812	2816	3.11	3508	3514	0.15	3530	3488	3661	3.64	3432	3487	1.58	1262	1214	3.89
La	324	328	322	1.59	76	76	0.89	72	63	70	3.04	111	113	1.79	92	88	4.66
Ce	192	190	187	2.41	150	153	1.90	141	131	145	2.76	207	197	4.82	198	189	4.46
Pr	69.08	73.31	70.64	4.92	18.98	18.89	0.47	17.74	16.10	17.76	0.10	27.85	27.91	0.20	23.14	22.64	2.21
Nd	261	289	279	8.20	78.96	77.67	1.64	72.80	66.41	74.07	1.73	119	120	0.49	95.84	90.97	5.21
Sm	52.42	54.54	53.79	3.28	16.21	16.21	0.00	15.27	14.29	15.34	0.47	25.80	26.13	1.30	19.06	18.49	3.05
Eu	7.42	7.44	7.89	5.73	6.37	6.90	8.02	6.56	6.35	7.22	9.49	8.06	8.77	8.43	6.00	5.36	11.3
Gd	56.58	60.66	60.00	6.06	16.41	17.38	5.76	15.78	14.75	16.05	1.68	26.39	27.44	3.91	20.03	18.56	7.59
Tb	8.24	8.80	8.68	5.58	2.41	2.43	0.91	2.29	2.15	2.31	0.59	3.82	4.02	5.12	2.79	2.62	6.49
Dy	48.02	51.09	49.94	5.09	13.85	13.88	0.18	13.19	12.58	13.32	0.94	21.61	21.90	1.36	15.89	15.32	3.65
Ho	9.47	9.93	9.82	4.10	2.71	2.70	0.40	2.59	2.46	2.66	2.92	3.98	4.18	5.01	3.24	2.92	10.1
Er	24.90	26.64	26.14	5.67	7.28	7.36	1.03	7.15	6.66	7.24	1.27	10.28	10.66	3.65	8.72	8.23	5.78
T	3.46	3.55	3.58	2.86	0.98	0.96	1.47	1.02	0.93	0.99	2.66	1.33	1.38	3.43	1.25	1.16	6.84
Yb	20.89	21.48	20.89	2.67	5.79	5.76	0.57	6.63	6.15	6.28	5.54	7.86	7.88	0.24	7.75	7.56	2.41
Lu	3.02	3.22	3.01	6.20	0.86	0.82	4.05	0.98	0.89	0.96	2.42	1.12	1.13	1.15	1.18	1.12	5.73
Hf	16.98	17.59	17.24	2.90	25.82	24.91	3.58	25.79	23.95	25.19	2.34	30.18	30.93	2.44	14.00	13.42	4.23
Ta	2.82	2.63	2.73	5.88	2.27	1.90	17.93	2.19	2.02	2.18	0.42	2.17	2.33	6.95	3.31	3.54	6.87
Pb	23.74	26.56	21.53	17.21	33.69	34.55	2.53	46.41	39.96	50.00	7.44	30.32	29.19	3.77	14.34	13.23	8.05
Th	14.67	16.34	13.36	16.50	10.12	9.64	4.84	11.55	10.15	11.36	1.69	7.56	6.69	12.22	4.71	4.50	4.50
U	5.69	5.89	5.75	3.00	2.60	2.53	2.51	2.57	2.26	2.50	2.71	2.14	2.21	3.34	1.35	1.31	2.72

Concentrations of samples digested by NH_4F (all new data presented in this paper) were calculated relative to a calibration curve, constrained using multiple gravimetric dilutions (0.1 ppb, 0.2 ppb, 0.5 ppb, 1 ppb, 2 ppb) of a 1 ppm stock standard solution comprising 57 elements. This procedure was undertaken to quantitatively calculate the concentrations of both standards run (GSP-2 and JR-1) and to confirm the accuracy of results. The 0.5 ppb calibration solution was measured after every four samples to monitor instrumental drift. Procedural blanks were <0.1 ppb for all elements, with most below detection limits. Abundances were calculated using the following equation:

$$\text{Sample } C_i = ((\text{Sample}_{\text{CPS}} \times c) / \text{Sample}_{\text{Dil}}) / 1000 \quad (3)$$

where c is the gradient of the calibration slope. Measured counts from calibration and unknown solutions were blank-corrected using the acid blank and procedural blank, respectively. Repeated analyses of rock standards GSP-2 and JR-1 yielded offsets from preferred values of $<10\%$ for all elements except Ho, Er ($<15\%$), Y ($<20\%$) and Zn ($>20\%$), and 2s.d.% similarly of $<10\%$ for all elements (Table S4; Electronic Appendix 4). BCR-2 was digested using both the HF and NH_4F methods in order to check for consistency. Measurements of this solution using NH_4F gave offsets from preferred values of $\leq 8\%$ for all elements except Mn and Ni ($<20\%$), comparable to, if not better than those using the HF technique (Table S3).

Sample preparation for isotope ratio measurements

Samples were selected for isotopic analysis by Thermal Ionisation Mass Spectrometry (TIMS) to cover the range of compositions measured by XRF and ICP-MS. Separation methodologies for Sr, Nd and Pb were based on those of Pin *et al.* (2014), with minor modification where appropriate (Fig. B). Rock powders were digested in pre-cleaned Savillex screw-topped Teflon beakers using the HF and NH_4F digestion techniques detailed above for ‘mafic’ (<65 wt % SiO_2) and ‘silicic’ (>65 wt% SiO_2) samples, respectively. Two aliquots of the final solution, one for Sr and Nd (containing ≥ 200 ng Nd), and one for Pb (containing ≥ 200 ng Pb) were taken and transferred into pre-cleaned beakers. Solutions were dried ready for processing.

Prior to element-specific columns, ‘silicic’ Sr and Nd solutions were passed through pre-cleaned 2ml Biorad™ columns filled with AG50W-X8 cation resin in order to pre-

concentrate the Sr and reduce the matrix load for the subsequent columns. Pre-concentration was achieved by loading the sample in 1 ml of 2.5M HCl, then eluting to waste. One ml then 6 ml of 2.5 M HCl. Sr was then collected in 10 ml of 2.5 M HCl. A further 7 ml of 2.5 M HCl was eluted to waste, then 10 ml of 6 M HCl was added to collect the Nd-bearing fraction. All fractions were then dried. The Sr-bearing fractions were taken up in 9 M HNO₃ to ensure conversion to nitrate form prior to being dried again in preparation for further purification. The ‘mafic’ Sr and Nd aliquots did not require pre-concentration on the cation column.

Table S4: Accuracy and precision of analyses (n=2) of standards analysed during ICP-MS runs of samples prepared using NH₄F digestion techniques.

	GSP-2					JR-2				
	Preferred	Average	Offset	2 s.d.	2 s.d.%	Preferred	Average	Offset	2 s.d.	2 s.d.%
Li	36	35.45	-1.52	0.90	2.53	61.4	64.38	4.85	1.35	2.10
Sc	6.3	6.38	1.29	0.35	5.43	5.07	5.17	2.05	0.45	8.69
V	52	51.12	-1.69	1.94	3.80	7	5.44	-22.22	0.08	1.41
Cr	20	18.97	-5.13	0.84	4.43	2.83	1.26	-55.60	0.70	55.71
Co	7.3	6.99	-4.19	0.25	3.51	0.83	0.55	-33.73	0.00	0.04
Ni	17	15.33	-9.82	1.11	7.23	1.67	0.37	-77.83	0.04	10.64
Cu	43	40.12	-6.71	2.76	6.87	2.68	1.78	-33.76	0.30	16.76
Zn	120	94.96	-20.87	7.50	7.90	30.6	23.70	-22.54	3.27	13.82
Ga	22	21.33	-3.04	0.67	3.16	16.1	16.15	0.30	1.38	8.52
Rb	245	233.38	-4.74	9.14	3.91	257	247.10	-3.85	20.60	8.34
Sr	240	218.14	-9.11	8.44	3.87	29.1	26.00	-10.67	1.68	6.45
Y	28	23.38	-16.50	0.81	3.44	45.1	37.72	-16.37	2.61	6.91
Zr	550	593.91	7.98	9.33	1.57	99.9	88.57	-11.34	7.21	8.14
Nb	27	26.82	-0.65	1.14	4.24	15.2	15.61	2.69	1.06	6.77
Cs	1.2	1.11	-7.72	0.05	4.87	20.8	19.21	-7.63	1.44	7.50
Ba	1340	1317.63	-1.67	29.59	2.25	50.3	44.37	-11.79	2.80	6.30
La	180	177.46	-1.41	4.52	2.55	19.7	18.45	-6.33	1.11	6.03
Ce	410	421.21	2.73	3.02	0.72	47.2	44.91	-4.85	2.19	4.87
Pr	51	53.22	4.35	1.37	2.58	5.58	5.62	0.77	0.27	4.79
Nd	200	193.82	-3.09	3.41	1.76	23.3	21.82	-6.36	1.09	5.01
Sm	27	24.87	-7.89	0.59	2.35	6.03	5.58	-7.47	0.43	7.72
Eu	2.3	2.50	8.89	0.17	6.59	0.3	0.28	-8.28	0.02	6.21
Gd	12	18.83	56.88	2.91	15.48	5.06	5.46	7.82	0.48	8.79
Tb		1.79		0.16	9.21	1.01	0.99	-1.80	0.07	6.65
Dy	6.1	5.51	-9.59	0.08	1.41	5.69	5.96	4.81	0.30	5.03
Ho	1	0.89	-10.87	0.04	4.46	1.11	1.23	10.90	0.05	3.77
Er	2.2	3.05	38.83	0.18	6.01	3.61	4.06	12.40	0.24	5.97
Tm	0.29	0.27	-5.34	0.01	2.85	0.67	0.63	-5.90	0.03	5.27
Yb	1.6	1.70	6.11	0.07	3.94	4.55	4.37	-3.98	0.24	5.57
Lu	0.23	0.26	12.76	0.00	1.92	0.71	0.72	1.17	0.02	2.30
Hf	14	15.24	8.84	0.10	0.65	4.51	4.61	2.27	0.32	6.88
Ta		1.02		0.02	2.06	1.86	1.90	1.88	0.07	3.70
Pb	42	44.70	6.42	4.60	10.30	19.3	16.78	-13.05	0.83	4.97
Th	105	101.62	-3.22	0.61	0.60	26.7	24.79	-7.14	0.68	2.74
U	2.4	2.37	-1.43	0.06	2.65	8.88	8.32	-6.26	0.36	4.33

Concentrations in ppm. Preferred values from Georem (<http://georem.mpch-mainz.gwdg.de/>) for GSP-2 and Imai *et al.* (1995) for JR-1. Full standard data given in Electronic Appendix 3.

For Sr separation, all fractions were taken up in 6M HNO₃ ready to pass through cleaned columns loaded with ~100 µl pre-cleaned Sr-Spec resin. The Sr-Spec resin was pre-conditioned with ~0.5 ml of 9M HNO₃. The sample was loaded before sequential elution to waste of 100, 200 and 3 x 1000 µl of 9M HNO₃ which removed Rb, Ba, Ca and other matrix elements. For ‘mafic’ samples, these elutions were collected for further separation of Nd. Two ml of 0.05 M HNO₃ was then passed through the Sr-Spec column to back extract the Sr.

For ‘mafic’ samples, the dried Nd fraction was taken up with 1ml of 1 M HNO₃ containing 50 mg ml⁻¹ ascorbic acid (C₆H₈O₆) in order to reduce the Fe_(III) to Fe_(II) because the oxidation state of the Fe has been shown to be important in obtaining good yields for the REE on TRU-Spec columns (Horwitz *et al.*, 1993; Pin & Zaldegui, 1997). To reduce the blank inherent in the ascorbic acid, the nitric-ascorbic acid solution was pre-cleaned by passing it through a TRU-Spec column. Reduction of the Fe after addition of the ascorbic was accompanied by a colour change - when the sample solution turned from yellow to grey-brown, the solution was ready for column separation. The TRU-Spec columns were pre-conditioned with 0.25 ml of the HNO₃-ascorbic solution before addition of the sample solution. Then 50, 100, 200 and 2 x 1000 µl of 1M HNO₃ were eluted to waste. Then a LREE-, Th- and U-bearing fraction was back extracted in 2 x 1 ml of 0.05 HNO₃, then dried. The pre-concentrated ‘silicic’ samples did not require pre-concentration on TRU-Spec columns, having already been pre-concentrated on the cation column.

For final chromatographic separation of Nd from other REE, dried fractions were taken up in 300 µl of 0.25 M HCl and passed through LN-Spec columns that had been pre-cleaned with 6 M HCl (three times) before being pre-conditioned with 0.25 M HCl (twice). The lanthanides were sequentially separated through further additions of 0.25 M HCl. Column calibrations were undertaken to identify position of the Nd fraction in the elution profile, and to minimise Ce and Pr overlap.

The calibration of LN-Spec columns was carried out by collecting 400 µl increments of 0.25 M HCl until 22 ml of 0.25 M HCl had been passed through the column. A subsequent 1 ml addition of 6 M HCl was also collected. These 400 µl increments were dried and taken up in 1 % HNO₃ for semi-quantitative analysis on the Element 2. The peak in Nd counts in the column increments indicated that collection of the 8 to 12 ml 0.25 M fraction produced good separation for Nd and contained no detectable Sm (Electronic Appendix 3).

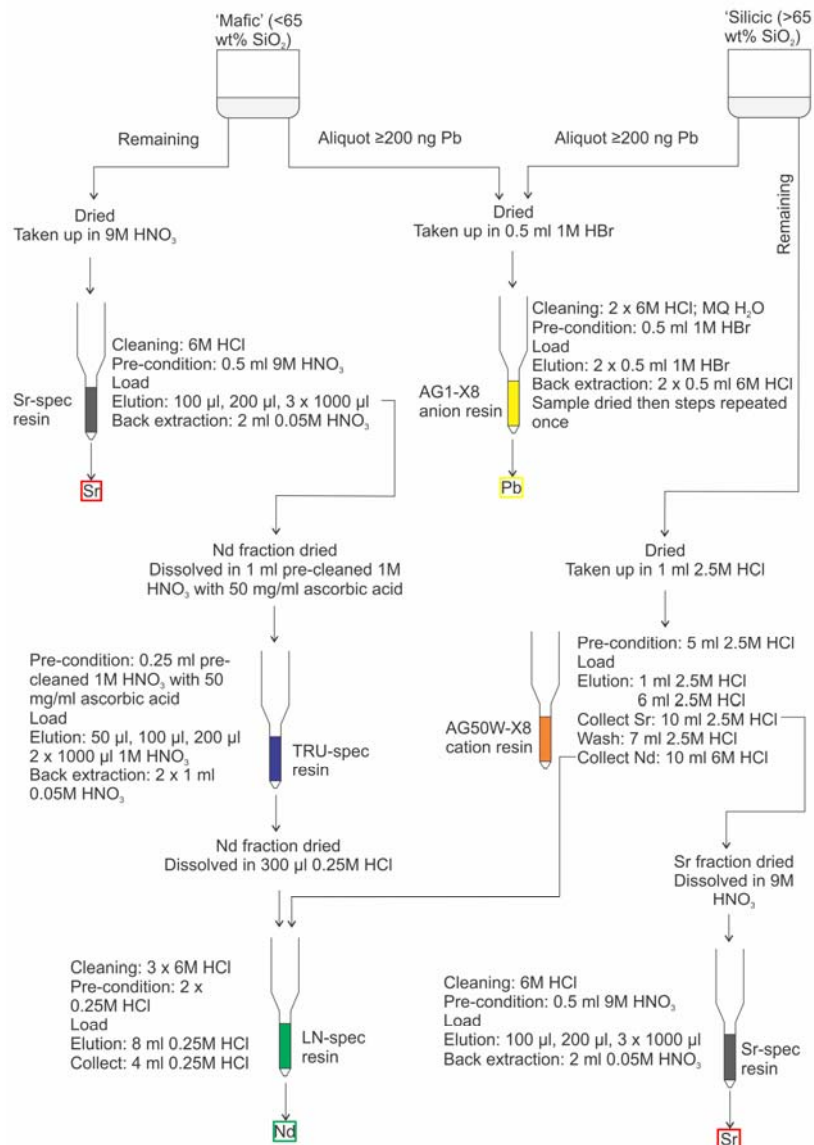


Fig. B: Schematic diagram showing the procedure for separation of Sr, Nd and Pb for isotopic analysis by extraction chromatography, based on the method of Pin *et al.* (2014).

Separation of Pb was achieved using the traditional ‘bromide separation’ approach on AG1-X8 anion resin (e.g. Strelow & von Toerien, 1966; Oversby, 1975). The pre-cleaned resin was transferred to columns made from 1 ml pipette tips, then finally cleaned by passing MQ water then 6M HCl through the resin twice, then residual HCl was washed out with ~2.5 ml of MQ H₂O. The resin was pre-conditioned with 0.5 ml 1M HBr and the sample loaded in 0.5 ml 1M HBr. Two passes of 0.5 ml 1M HBr were eluted to waste, then the Pb was back-extracted in two 0.5 ml washes of 6M HCl. Approximately 0.05 ml conc. HNO₃ was added to

the collected solution to exsolve the remaining Br and the solution taken to dryness. The sample was then taken up once again in 0.5 ml of 1M HBr and the entire process repeated.

Isotopic ratio mass spectrometry

Sr, Nd and Pb isotope ratios were measured using a ThermoFisher Triton Thermal Ionisation Mass spectrometer (TIMS) first at the Open University, UK (by B. L. A. Charlier), then at Victoria University of Wellington (VUW), New Zealand (by E. J. Swallow), using the same instrument.

The dried Sr fractions were dissolved in ~1 µl concentrated HNO₃ and loaded with 0.7 µl of pre-cleaned TaF₂ activator (Birck, 1986) on to outgassed, zone refined single Re filaments using the methods of Charlier *et al.* (2006). The samples were added to the filament before the activator had completely dried to ensure the load was mixed on the filament, then glowed to a dull red. For analysis, filaments were heated slowly to 1450-1500 °C in order to generate ion beams of ~50 mV on ⁸⁴Sr (i.e. 7 to 8 V on ⁸⁸Sr), and measurements consisted of 240 ratios measured in blocks of 10 with an integration time of 16.777 s for each ratio measured. A peak-centre and tune was automated at the beginning of every 10 blocks and a 1 min baseline was measured at half mass at the beginning of every block (and rotation of the amplifiers). Measurements were fractionation corrected using the exponential law to ⁸⁶Sr/⁸⁸Sr = 0.1194, and Rb interference was corrected by measuring ⁸⁵Rb and applying a correction using ⁸⁷Rb/⁸⁵Rb = 0.385707 (Rosman & Taylor, 1998). Repeated analyses of NBS987 yield an average of 0.710255 ± 0.0000013 (2 s.d.; n = 25). Isotopic ratios from different runs were normalised to a value of 0.71025 for NBS987 (Thirlwall, 1991) to ensure internal consistency. Sr procedural blanks were 110 and 200 pg for the ‘mafic’ and ‘silicic’ samples, respectively, insignificant compared to the ~1000 ng sample load.

The dried Nd fractions (200 – 800 ng) were taken up in 1-2 µl conc. HCl then loaded onto the evaporation side of an outgassed, zone refined double Re filament assembly following the addition, and drying, of 1 µl of 0.1M H₃PO₄ loading solution. A further 1 µl of loading solution was then added and dried. The filament was then turned up to a dull red glow for a few seconds. Nd was run with an evaporation filament (containing the sample) and a hotter ionisation filament (1650-1800 °C) to generate 2-8 V on ¹⁴²Nd depending on the amount of sample loaded. 270 ratios, each with an integration time of 8.389 s, were measured in blocks of 10 with a baseline measured at half mass before each block. A peak-centre and tune was completed after each block of 10 ratios. Ratios were fractionation corrected using the

exponential law to $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$, and any presence of Ce and Sm corrected using values of $^{140}\text{Ce}/^{142}\text{Ce} = 0.20667$ and $^{144}\text{Sm}/^{147}\text{Sm} = 7.97297$ (Rosman & Taylor, 1998). Repeated analyses of standards J&M (internal standard) and La Jolla (international standard) yield values of $^{143}\text{Nd}/^{144}\text{Nd} = 0.511818 \pm 0.000004$ (2 s.d.; $n = 26$) and 0.511845 ± 0.000002 (2 s.d.; $n = 6$), respectively, similar to the long term laboratory average (J&M: 0.511821 ± 0.000002 2 s.d.) and the value of Thirlwall (1991) for La Jolla (0.511856 ± 0.000007 2 s.d.). Procedural blanks were 8 and 300 pg for the ‘mafic’ and ‘silicic’ samples, respectively which can be considered negligible compared to the sample amount loaded onto the filament (200–800 ng).

The dried Pb fraction was taken up in 2 μl of H_3PO_4 -silica gel mix (Gerstenberger & Haase, 1996). Pb was analysed using a double-spike methodology (Thirlwall, 2000). Half the sample (approximately 150 ng), was loaded onto single outgassed, zone refined Re filaments and measured for 180 ratios at 8.389 second integrations. A 1 μl aliquot of $^{207}\text{Pb}/^{204}\text{Pb}$ spike (Thirlwall, 2000) per 100 ng Pb was then admixed to the remaining sample, which was then loaded and measured as before for 120 ratios. Spiked and natural samples were run during separate analytical sessions, on filaments designated for spiked and natural samples only in order to prevent an cross-contamination from residual spike from previous runs. Loading was carried out in an identical way to avoid the possibility of differential blank contributions to the spiked and natural runs. The measured data were deconvolved using a modified Newton-Raphson inversion to determine the isotope ratios without recourse to internal normalisation, and the uncertainties determined using a 5000 iteration Monte-Carlo simulation. Values for NBS981 were $^{206}\text{Pb}/^{204}\text{Pb} = 16.945 \pm 0.001$, $^{207}\text{Pb}/^{204}\text{Pb} = 15.503 \pm 0.001$ and $^{208}\text{Pb}/^{204}\text{Pb} = 36.737 \pm 0.004$ (2 s.d.; $n = 32$). Isotopic measurements from different runs were normalised to the NBS981 values of Todt *et al.* (1996). The procedural blanks for Pb were 10 pg (‘mafic’ samples) and 2 ng (‘silicic’). Even the higher blank content for the ‘silicic’ samples is considered insignificant when compared to the >200 ng of sample analysed.

IN-SITU GLASS AND MINERAL ANALYSIS

Glass and crystals were hand-picked from the preferred size fractions, mounted onto double-sided adhesive tape and cast in Epofix™ resin within 25 mm rounds and polished. Crystals were orientated parallel to the c-axis where identifiable and polished down to expose the mid-section. Chips of the basal ignimbrite vitrophyre were mounted in epoxy, and some pumice samples prepared as polished thin-sections.

Electron Probe Micro-Analysis (EPMA): major and minor elements

Major and minor elements were determined by electron probe micro-analysis (EPMA) using a JXA 8230 Superprobe at VUW. A ~25 nm carbon film was applied to samples prior to analysis. Backscattered electron images were made of all samples and qualitative and quantitative analysis was achieved using an energy dispersive spectrometer and five wavelength dispersive spectrometers (WDS), respectively. All analytical data in this study were determined using WDS. A ZAF correction (where Z is atomic number, A is absorption and F is fluorescence) was used to calculate all elemental concentrations (e.g. Carpenter, 2008). Calibration for quantitative analysis was undertaken using international standards of similar composition and structure to materials being analysed in order to reduce the effects of different matrices on the accuracy of the measurements. Count times were similar for both calibration standards and samples. Minor elements (<1 wt%) were calibrated against synthetic oxides. Calibration standards and secondary standards were also run as unknowns to check the precision and accuracy of each run and to monitor any instrumental drift.

Glass was analysed by EPMA using a 15 kV accelerating voltage and a defocused 20 μm spot size. Si, Al, Na, K, Fe Ca were analysed using a 2 nA sample current, as recommended by Humphreys *et al.* (2006), to minimise alkali migration. Electron-beam effects were further reduced by the analysis of Si and Na first. Subsequently, Ti, Mg and Mn were analysed using an 8 nA current. Count times (on peak/background) were 15/8 (Na), 30/15 (Si, Al), 40/20 (K, Ca, Ti, Mg) and 60/20 s (Fe, Mn). A 15 kv accelerating voltage was used for the analysis of all crystal phases with a sample current of 12 nA and a spot size of ~1 μm . Feldspars and pyroxenes were analysed with 30 s count time on peak and a 15 s background count for all elements. For olivines, Ti, Si, Mg, Fe and Al were analysed with a 30 s count time on peak and a 15 s background count, whereas Ca, Mn and Ni were analysed over 60 and 30 s to improve counting statistics. Accuracy and precision of repeated standard analysis are given in Table S5.

Laser ablation (LA-ICP-MS): trace elements

Following determination of major and minor element concentrations by EPMA, glassy material was analysed for trace elements by Laser-Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) using a RESolution S155-SE 193nm Excimer Laser coupled to an Agilent 7500CS ICP-MS at VUW.

LA-ICP-MS analysis of glass was conducted using a 35 μm spot size which was placed coincident with the EPMA spot. Each analysis was preceded by two 70 μm diameter cleaning pulses followed by a 20 s pause to allow counts to return to background levels. Analyses consisted of a 30 s ablation at 5 Hz with a laser energy $\sim 5 \text{ mJ/cm}^2$. Each ablation was followed by a 15 s washout to allow a return to background. As some doubt has been expressed regarding the homogeneity of NIST synthetic glasses (e.g. Eggins & Shelley, 2002), a synthetic basaltic glass GSD-1G was used for calibration (Guillong *et al.*, 2005). ^{29}Si concentrations, derived from EPMA results for each sample, were used to internally standardise the LA-ICP-MS data. This is preferable to other major elements due to lower uncertainty of EPMA results for Si. BHVO-2G, BCR-2G and NIST 612 were analysed as secondary standards to cover a wide range in concentrations for each element and allow propagation of measurement uncertainties. Data were processed using LaserTRAM software (Loewen & Kent, 2012).

The mass spectrometer was tuned for optimal sensitivity by running a 50 μm raster on NIST 612 and simultaneously adjusting the torch and lens positions and gas flows. Pulse/analog (P/A) conversion factors were measured prior to every analytical session to ensure correct adjustment of the ICP-MS when switching between low (P) and high (A: $>10^6$ cps) counting modes. Five unknowns and a secondary standard (alternating NIST 612, BCR-2G and BHVO-2G) were bracketed by a calibrating standard (GSD-1G). Offsets for NIST 612 from preferred values (Jochum *et al.*, 2011) are $\leq 9 \%$ for all elements except Y, Zr, Nd ($<12 \%$) and Er, Gd ($<15\%$). 2 s.d.% precisions are better than 13 % except for Li, Zn ($<20 \%$) and Ti ($<30 \%$). Full standard data are detailed in Table S6.

Table S5: Accuracy and precision of repeated analyses of standard materials by EPMA (data in wt%).													
	Analysis	SiO₂	TiO₂	Al₂O₃	FeOt	MnO	MgO	CaO	Na₂O	K₂O	BaO	NiO	Cr₂O₃
Rhyolitic glass (VG-568) n=348 Jarosewich et al. (1980)	Preferred	76.96	0.08	12.17	1.08	0.02	0.03	0.45	3.52	4.93			
	Average	77.32	0.12	12.18	1.21	0.07	0.07	0.50	3.45	4.73			
	Offset	0.47	54.93	0.05	11.69	251.67	138.24	11.05	-2.06	-4.02			
	2 s.d.	1.53	0.18	0.46	0.23	0.12	0.11	0.11	0.36	0.47			
	2 s.d.%	1.98	148.52	3.77	18.92	177.02	155.45	21.62	10.40	9.87			
Orthoclase (Or-1A) n=460 Ingamells (1980)	Preferred	64.39		18.19	0.03				1.14	14.92	0.82		
	Average	64.42		18.30	0.01				1.14	14.95	0.79		
	Offset	0.05		0.58	-71.44				0.05	0.19	-3.44		
	2 s.d.	0.81		0.44	0.02				0.10	0.35	0.16		
	2 s.d.%	1.25		2.39	269.56				8.87	2.33	19.78		
Plagioclase (NMNH 115900) n=438 Jarosewich et al. (1980)	Preferred	51.25	0.05	30.91	0.46		0.14	13.64	3.45	0.18	0.05		
	Average	51.15	0.03	29.36	0.45		0.14	13.68	4.03	0.12	0.01		
	Offset	-0.19	-39.36	-5.02	-2.23		3.02	0.32	16.78	-31.78	-71.95		
	2 s.d.	0.92	0.03	0.95	0.17		0.03	0.52	0.42	0.08	0.04		
	2 s.d.%	1.79	88.83	3.24	37.95		19.68	3.79	10.52	65.42	276.96		
Scapolite P.S.U n=107 Ingamells (1983)	Preferred	49.78		25.05	0.17			13.58	5.20	0.94			
	Average	50.01		25.07	0.10			13.49	5.18	0.94			
	Offset	0.45		0.10	-43.67			-0.67	-0.42	0.01			
	2 s.d.	0.72		0.23	0.06			0.40	0.31	0.03			
	2 s.d.%	1.45		0.93	62.68			2.94	6.00	3.42			
Olivine (Springwater 2566) n=114 Jarosewich et al. (1980)	Preferred	38.95			16.62	0.30	43.58						
	Average	38.96		0.00	16.82	0.28	43.55						
	Offset	0.04			1.19	-8.12	-0.08						
	2 s.d.	0.26		0.01	0.59	0.17	0.37						
	2 s.d.%	0.66		326.33	3.53	62.96	0.86						

Table S5, continued													
	Analysis	SiO ₂	TiO ₂	Al ₂ O ₃	FeOt	MnO	MgO	CaO	Na ₂ O	K ₂ O	BaO	NiO	Cr ₂ O ₃
Pyroxene (Px-1) n=197 Ingamells (1980)	Preferred	53.94	0.26	0.66	2.91	0.07	16.93	24.55	0.24				0.21
	Average	53.88	0.26	0.67	2.93	0.06	17.14	24.49	0.25				0.22
	Offset	-0.11	0.43	1.28	0.75	-	1.25	-0.25	4.85				4.79
	2 s.d.	1.17	0.06	0.12	0.60	13.23	0.04	0.78	0.10				0.12
	2 s.d.%	2.17	21.18	17.27	20.30	69.07	4.57	1.82	41.40				54.78
Ilmenite (NMNH96189) n=114 Jarosewich et al. (1980)	Preferred		45.70		46.54	4.77	0.31						
	Average		54.12		46.82	4.15	0.28						
	Offset		18.42		0.60	-	-8.99						
	2 s.d.		6.01		1.48	13.08	0.06						
	2 s.d.%		11.11		3.16	59.45	22.91						
Hypersthene (USNM746) n=83 Jarosewich et al. (1980)	Preferred	54.09	0.16	1.23	15.22	0.49	26.79	1.52	0.05	0.01			0.75
	Average	54.41	0.10	0.98	15.29	0.48	27.51	1.23	0.02	0.13			0.67
	Offset	0.59	-	-20.27	0.49	-1.51	2.70	-	-67.95	1239.38			-10.91
	2 s.d.	3.09	37.01	0.04	0.70	0.11	1.23	19.23	0.04	0.74			0.20
	2 s.d.%	5.67	41.66	22.78	4.60	23.51	4.47	23.08	245.12	551.65			29.65
Augite (Kakanui) n=82 Jarosewich et al. (1980)	Preferred	50.73	0.74	8.73	6.34	0.13	16.65	15.82	1.27				0.15
	Average	50.76	0.95	8.85	6.41	0.14	16.73	15.79	1.60				0.15
	Offset	0.06	28.12	1.41	1.03	5.39	0.50	-0.16	25.74				-0.70
	2 s.d.	0.40	0.10	0.48	0.65	0.05	0.31	0.35	0.55				0.05
	2 s.d.%	0.80	10.13	5.45	10.16	34.09	1.87	2.21	34.51				34.38

Table S6: Accuracy and precision of repeated analyses of standard materials by LA-ICP-MS.															
	BHVO-2G n=116					BCR-2G n=116					NIST 612 n=210				
	Pref.	Average	% Offset	2 s.d.	2 s.d.%	Pref.	Average	% Offset	2 s.d.	2 s.d.%	Pref.	Average	% Offset	2 s.d.	2 s.d.%
Li	4.4	4.19	-4.82	1.84	43.94	9	8.99	-0.14	2.60	28.90	40.2	40.49	0.71	5.34	13.20
Ti	16300	15867	-2.65	1890	11.91	14100	13439	-4.69	1545	11.50	44	42.72	-2.90	16.09	37.65
Mn	1317	1403	6.59	221.5	15.79	1550	1679	8.32	272.7	16.24	38.7	40.90	5.70	5.16	12.61
Zn	102	114.9	12.60	14.95	13.02	125	153.2	22.52	18.45	12.04	39.1	38.23	-2.22	7.00	18.30
Ga	22	20.80	-5.45	2.53	12.17	23	22.31	-2.98	2.77	12.43	36.9	36.58	-0.88	3.41	9.33
Rb	9.2	8.88	-3.44	1.18	13.23	47	46.14	-1.82	3.97	8.60	31.4	30.83	-1.81	2.49	8.07
Sr	396	386.9	-2.30	43.68	11.29	342	341.4	-0.18	32.82	9.61	78.4	82.35	5.04	7.01	8.51
Y	26	24.29	-6.57	3.20	13.17	35	34.66	-0.97	4.29	12.38	38.3	42.64	11.33	3.94	9.24
Zr	170	163.3	-3.94	19.17	11.74	184	183.4	-0.31	20.78	11.33	37.9	41.87	10.49	4.20	10.04
Nb	18.3	16.79	-8.23	2.19	13.06	12.5	11.71	-6.32	1.39	11.89	38.9	40.31	3.63	3.27	8.12
Mo	3.8	4.11	8.20	1.83	44.59	270	265.6	-1.61	23.90	9.00	37.4	38.28	2.36	4.35	11.36
Ba	131	123.4	-5.79	15.86	12.85	683	668.5	-2.12	77.36	11.57	39.3	39.83	1.35	5.19	13.02
La	15.2	14.68	-3.39	1.85	12.58	24.7	25.15	1.83	3.01	11.97	36	38.75	7.65	3.68	9.50
Ce	37.6	36.36	-3.30	4.44	12.22	53.3	53.02	-0.53	6.36	11.99	38.4	40.69	5.95	3.34	8.22
Pr	5.35	4.93	-7.85	0.77	15.55	6.7	6.58	-1.84	0.92	14.06	37.9	40.15	5.92	3.28	8.17
Nd	24.5	23.76	-3.00	4.06	17.10	28.9	28.87	-0.11	4.54	15.74	35.5	39.06	10.04	4.26	10.90
Sm	6.1	5.77	-5.33	1.80	31.23	6.59	6.50	-1.29	1.65	25.39	37.7	40.72	8.02	4.69	11.51
Eu	2.07	2.01	-2.70	0.57	28.18	1.97	1.97	-0.13	0.55	28.09	35.6	38.29	7.56	3.37	8.79
Gd	6.16	6.15	-0.17	1.80	29.34	6.71	6.82	1.71	1.97	28.83	37.3	42.82	14.80	5.40	12.60
Tb	0.92	0.87	-5.37	0.26	30.14	1.02	0.98	-3.93	0.30	30.32	37.6	40.06	6.55	3.96	9.89
Dy	5.28	4.94	-6.38	1.15	23.28	6.44	6.25	-2.97	1.32	21.19	35.5	38.70	9.00	4.42	11.43
Ho	0.98	0.94	-4.52	0.28	30.28	1.27	1.25	-1.57	0.33	26.72	38.3	40.70	6.26	3.94	9.67
Er	2.56	2.49	-2.61	0.76	30.36	3.7	3.71	0.14	0.96	25.89	38	42.66	12.25	4.96	11.62
Tm	0.34	0.31	-10.02	0.17	55.32	0.51	0.50	-1.81	0.22	44.50	36.8	39.32	6.84	3.88	9.87
Yb	2.01	1.84	-8.60	0.96	52.02	3.39	3.35	-1.31	1.18	35.31	39.2	41.63	6.21	5.13	12.33
Lu	0.28	0.26	-6.30	0.17	63.36	0.50	0.49	-3.49	0.25	51.87	37	39.82	7.62	4.28	10.75
Hf	4.32	4.18	-3.35	1.00	23.86	4.84	4.73	-2.23	1.13	23.78	36.7	38.66	5.35	4.15	10.72
Ta	1.15	1.01	-12.16	0.26	26.08	0.78	0.71	-8.88	0.23	32.11	37.6	39.14	4.09	3.84	9.80
Pb	1.7	1.80	5.79	0.57	31.89	11	10.96	-0.40	1.44	13.17	38.57	40.46	4.91	4.23	10.44
Th	1.22	1.19	-2.73	0.41	34.54	5.9	5.97	1.25	1.06	17.81	37.79	41.21	9.06	4.79	11.61
U	0.40	0.42	4.85	0.22	53.00	1.69	1.73	2.18	0.47	27.00	37.38	38.56	3.15	4.16	10.79
Preferred values from Georem (http://georem.mpch-mainz.gwdg.de/). Data in ppm.															

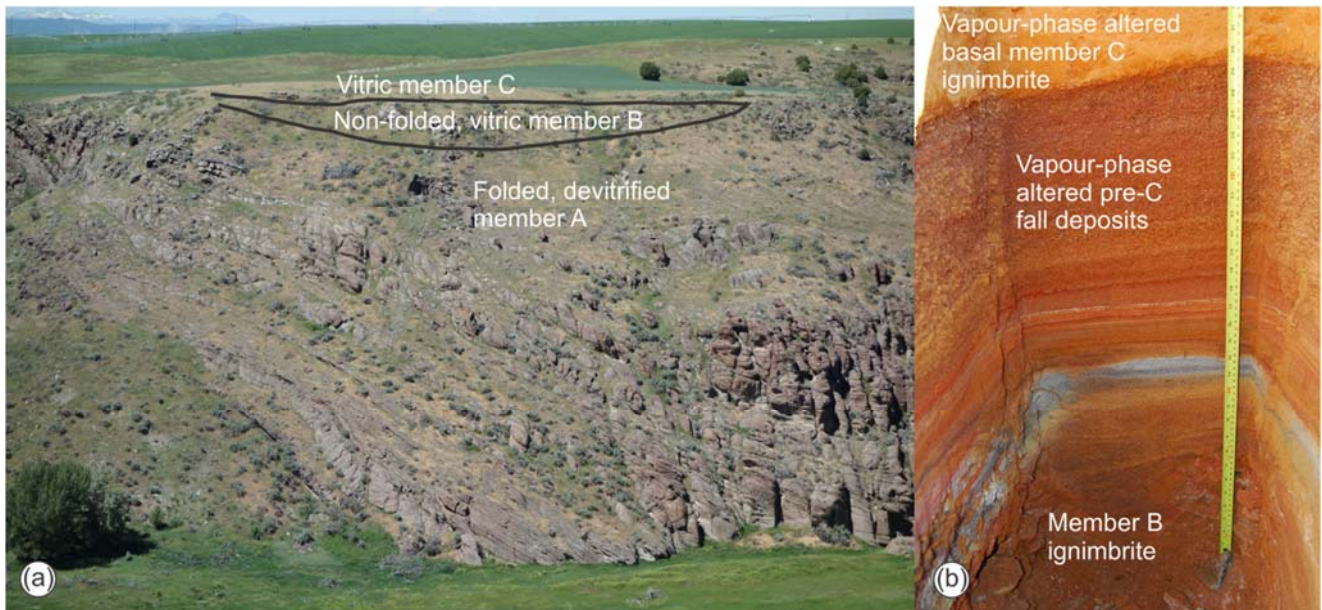


Fig. S1. Photographs showing evidence for time breaks within the HRT eruption.

(a): True right (north) bank of the Teton River about 2.5 km upstream of the site of the Teton Dam, showing rheomorphically folded and devitrified member A ignimbrite forming most of the gorge wall (cf. Geissman *et al.*, 2010). Overlying member B ignimbrite is little deformed and partly vitric, and infills shallow swales on the surface of the folded member A ignimbrite. These relationships indicate that member A had been deformed and its top surface cooled prior to deposition of member B, leading to an inference of a time gap of weeks to months between the two. Member C ignimbrite is wholly vitric in this area, implying a significantly longer time break between B and C.

(b): Photo of a pit dug through the pre-C fall deposits down to the contact with the upper member B ignimbrite. Despite the welding and cooling break between members B and C (see panel a), pre-C fall deposits and the base of member C ignimbrite are vapour-phase altered. This indicates that the combined material of members A and B was still degassing when fall the fall deposits and member C ignimbrite were emplaced, constraining the time gap to years to decades. A close modern comparison for estimating the time break is with the Valley of Ten Thousand Smokes ignimbrite (Katmai, Alaska), which is of comparable thickness, was welded, and where decay of vigorous high-temperature fumarolic activity occurred within 20-30 years (Hildreth & Fierstein, 2012).

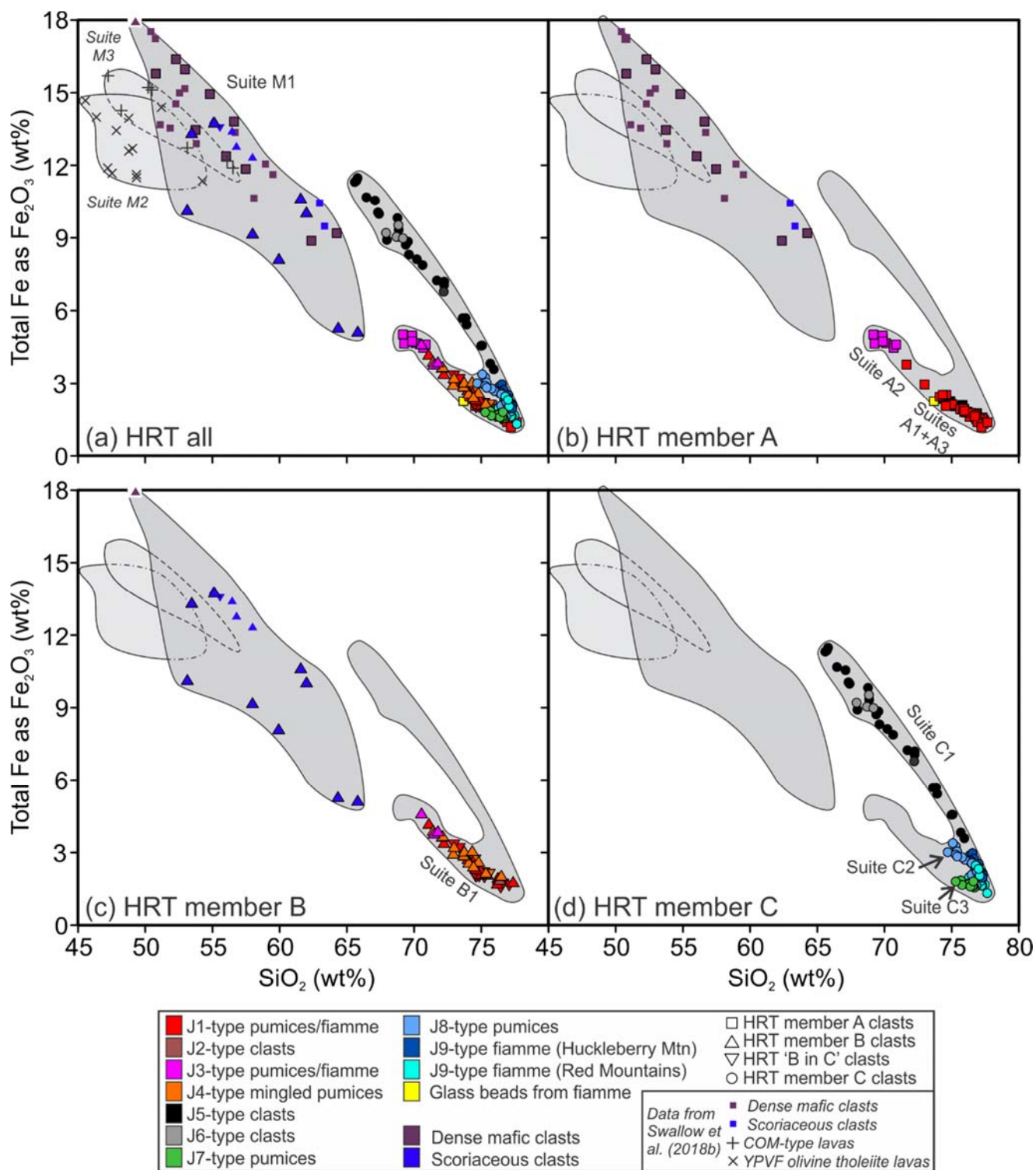


Fig. S2. Total Fe as Fe_2O_3 versus SiO_2 for samples analysed in this study for the entire HRT (and relevant regional mafic eruptives: Swallow *et al.*, 2018b) and individual members. Analytical 2 s.d. precisions are smaller than the symbol size. Other information as in Fig. 5 in the main paper. See Electronic Appendix 3 for the full data set.

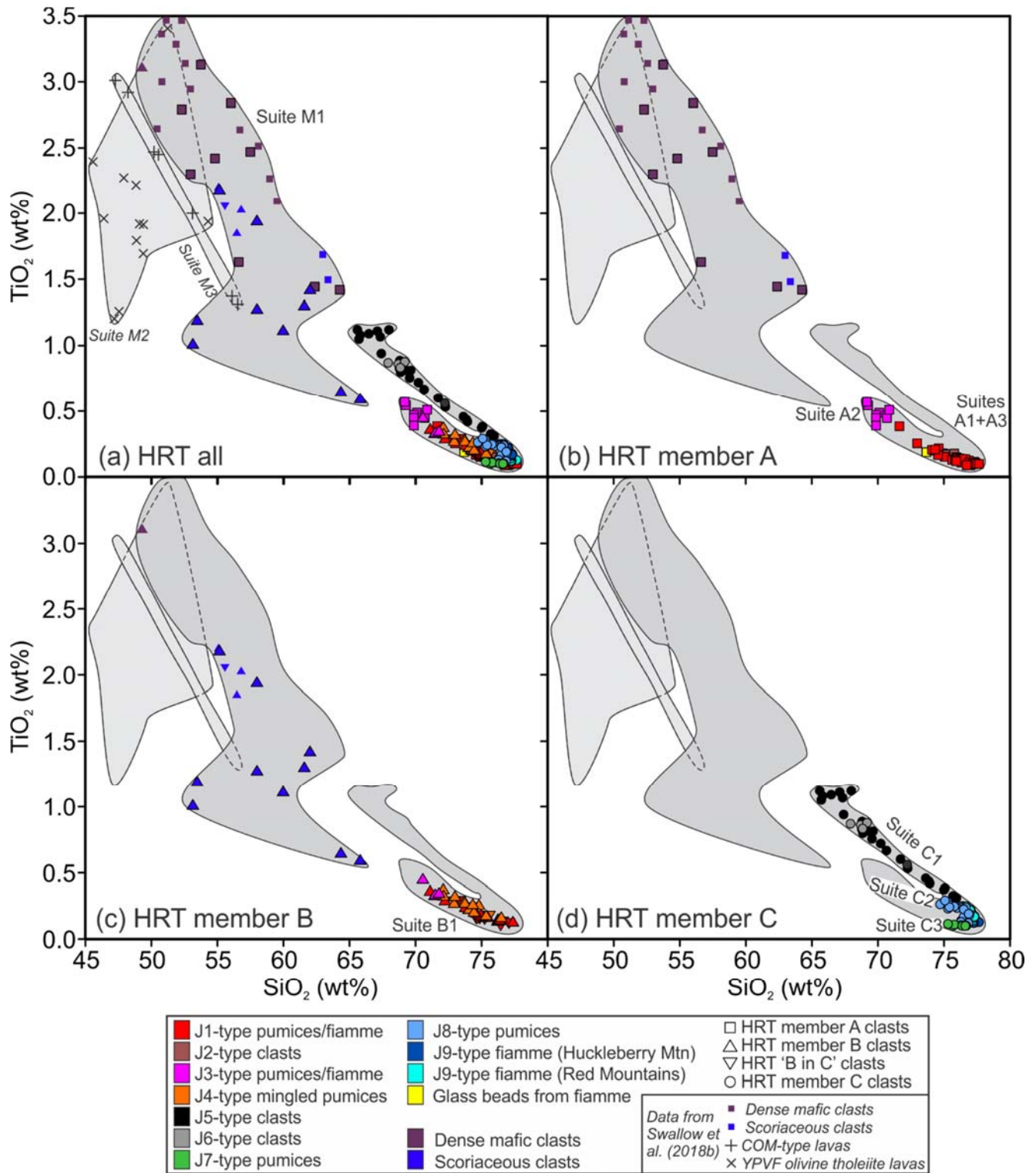


Fig. S3. TiO_2 versus SiO_2 for samples analysed in this study for the entire HRT (and relevant regional mafic eruptives: Swallow *et al.*, 2018b) and individual members. Analytical 2 s.d. precisions are smaller than the symbol size. Other information as in Fig. 5 in the main paper. See Electronic Appendix 3 for the full data set.

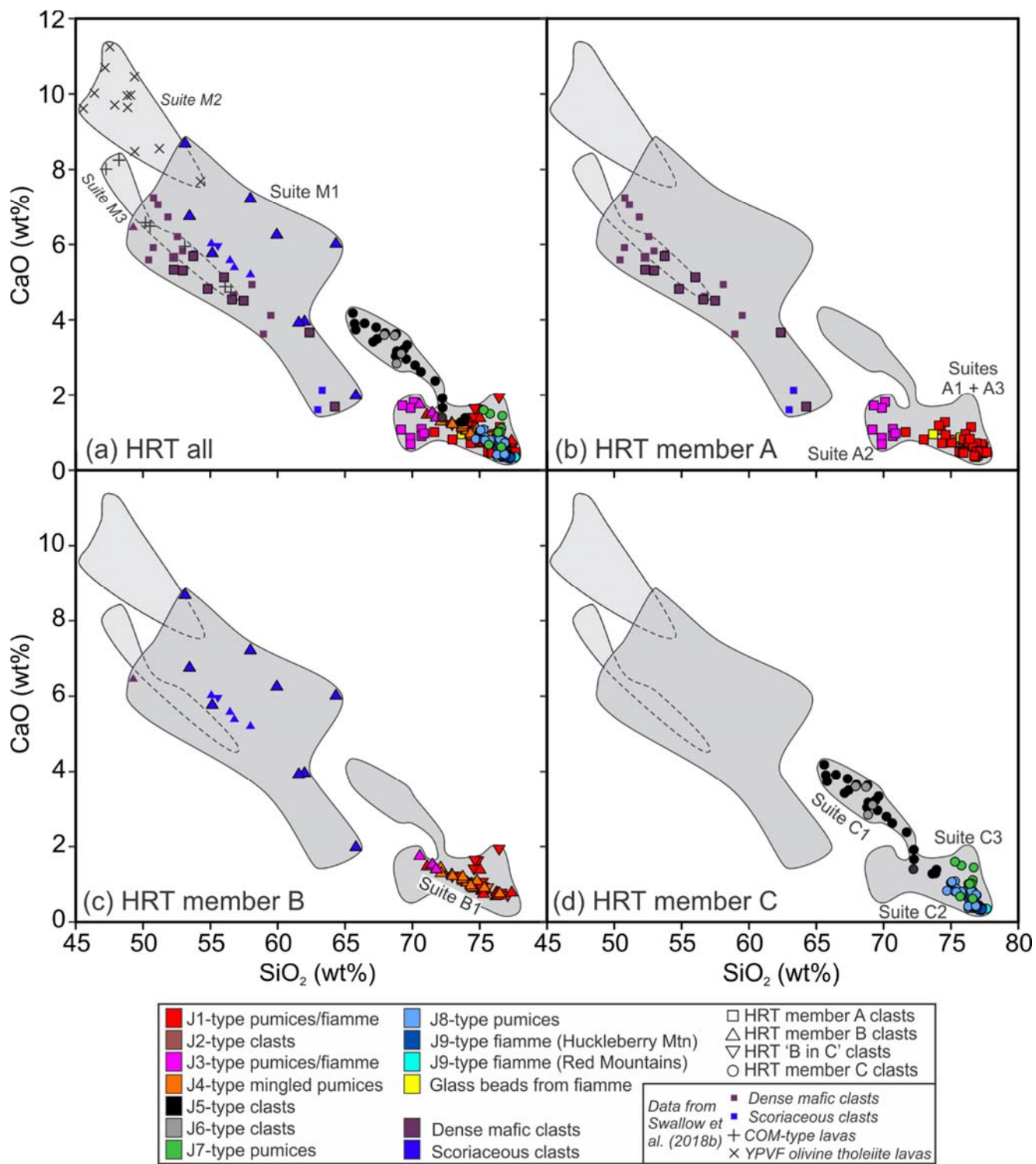


Fig. S4. CaO versus SiO₂ for samples analysed in this study for the entire HRT (and relevant regional mafic eruptives: Swallow *et al.*, 2018b) and individual members. Analytical 2 s.d. precisions are smaller than the symbol size. Other information as in Fig. 4 in the main paper. See Electronic Appendix 3 for the full data set.

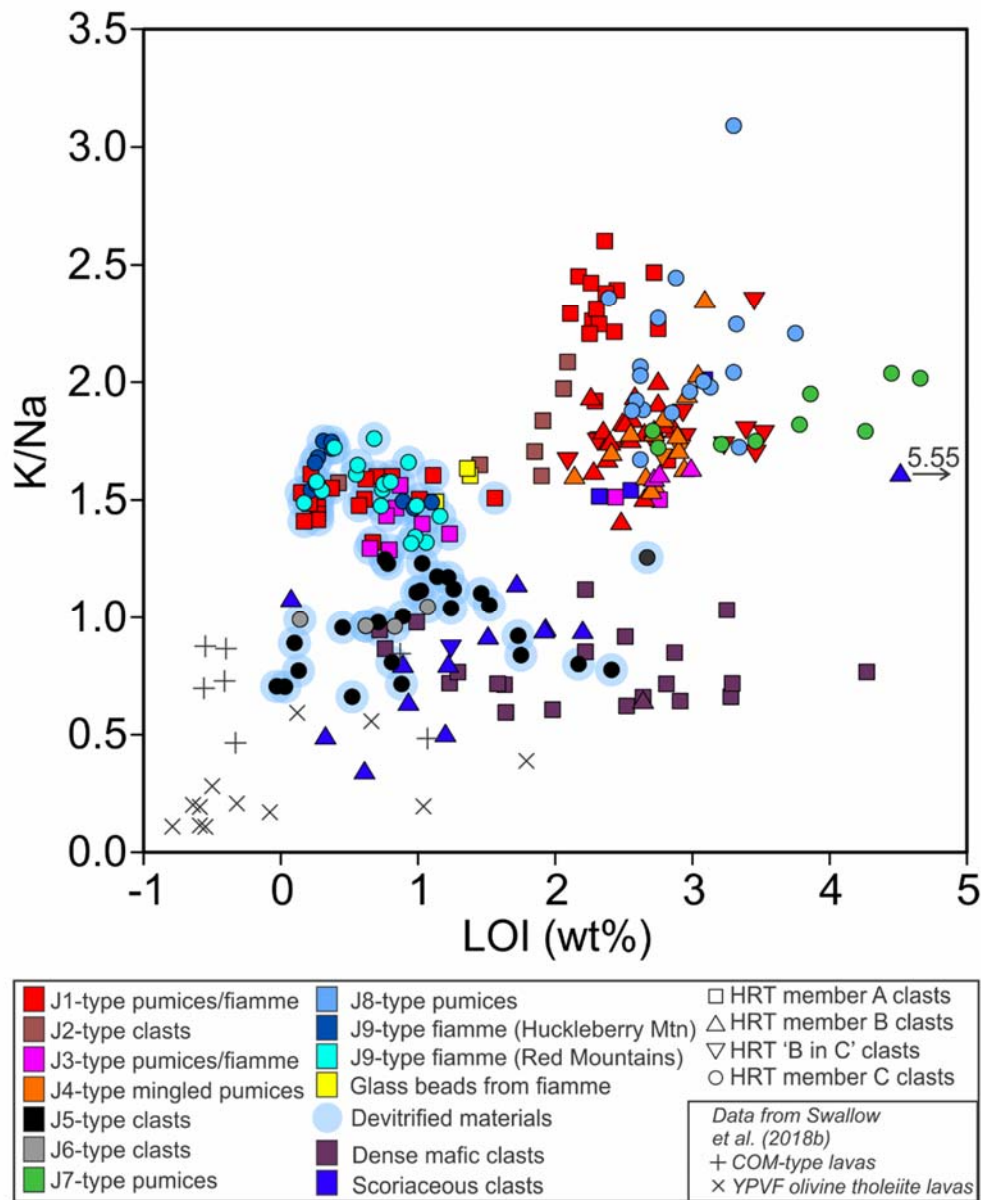


Fig. S5. K/Na values versus single-clast loss on ignition (LOI) for samples analysed in this study for the entire HRT (and relevant regional mafic eruptives: Swallow *et al.*, 2018b). Devitrified juvenile clasts are highlighted in blue. Although there is a consistent offset in LOI values between the glassy and crystalline materials, and broadly higher LOI values are seen in glassy pumices, there are overlapping K/Na values between different juvenile clast types and no clear relationships between LOI values and K/Na ratios in the rhyolitic components. The K/Na ratios would be expected to increase with hydration if alkali mobility (especially Na loss) were important (Hildreth and Wilson 2007). See Electronic Appendix 3 for the full data set.

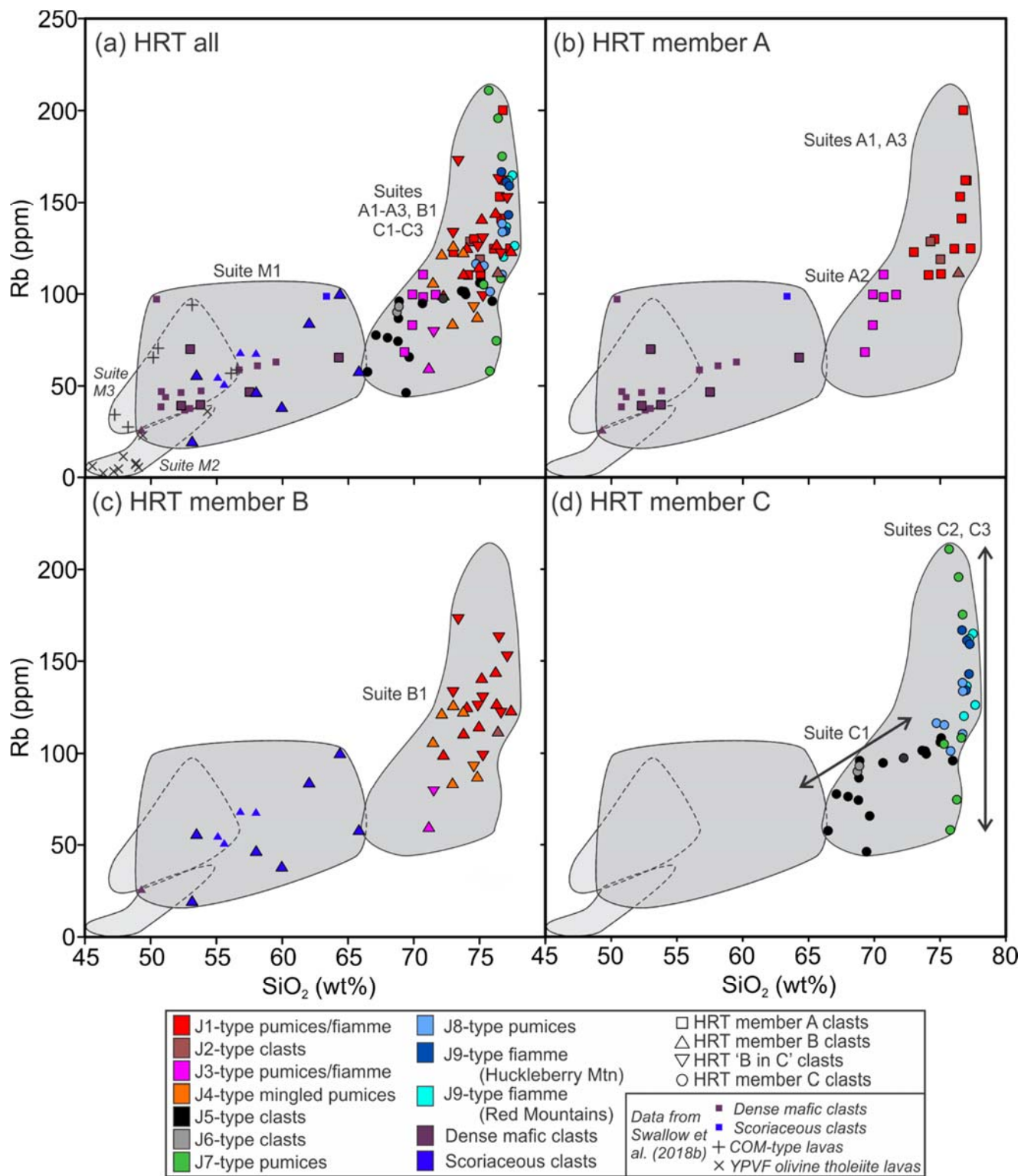


Fig. S6. Rb versus SiO₂ for samples analysed in this study for the entire HRT (and relevant regional mafic eruptives: Swallow *et al.*, 2018b) and individual members. Analytical 2 s.d. precisions are smaller than the symbol size. Other information as in Fig. 5 in the main paper. See Electronic Appendix 3 for the full data set.

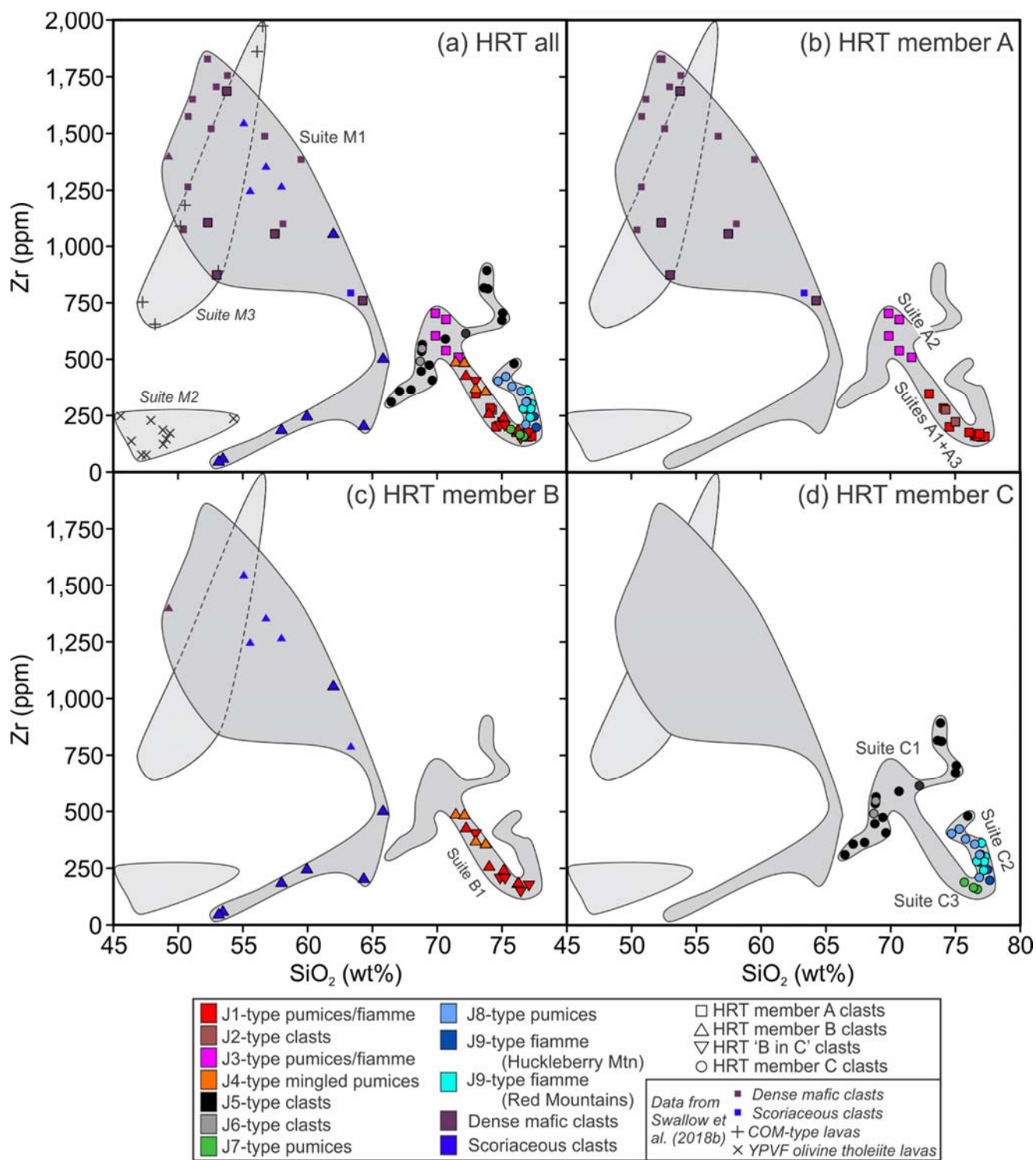


Fig. S7. Zr versus SiO₂ for samples analysed in this study for the entire HRT (and relevant regional mafic eruptives: Swallow *et al.*, 2018b) and individual members. Analytical 2 s.d. precisions are smaller than the symbol size. Other information as in Fig. 5 in the main paper. See Electronic Appendix 3 for the full data set.

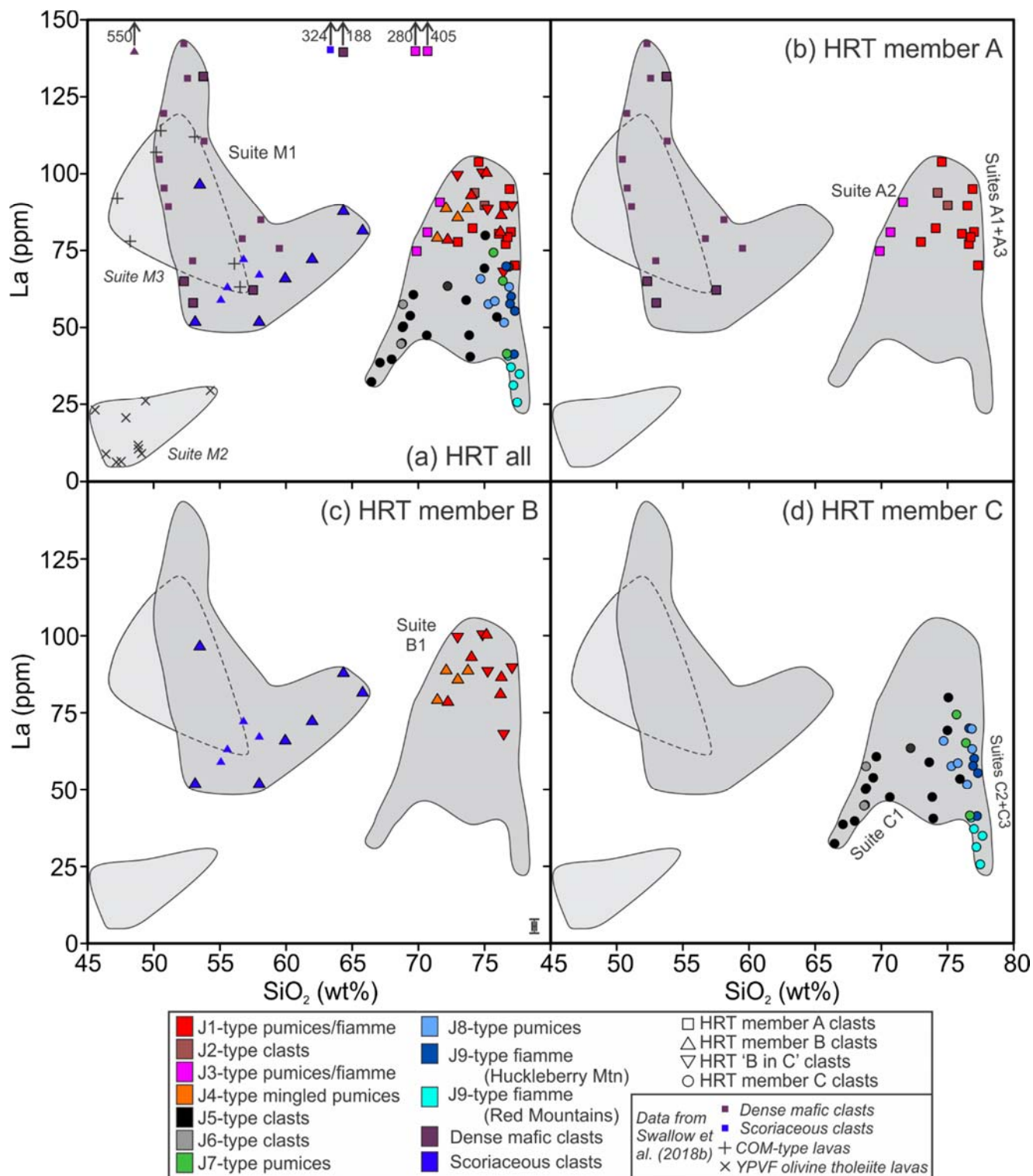


Fig. S8. La versus SiO₂ for samples analysed in this study for the entire HRT (and relevant regional mafic eruptives: Swallow *et al.*, 2018b) and individual members. Analytical 2 s.d. precisions are smaller than the symbol size. Other information as in Fig. 5 in the main paper. See Electronic Appendix 3 for the full data set.

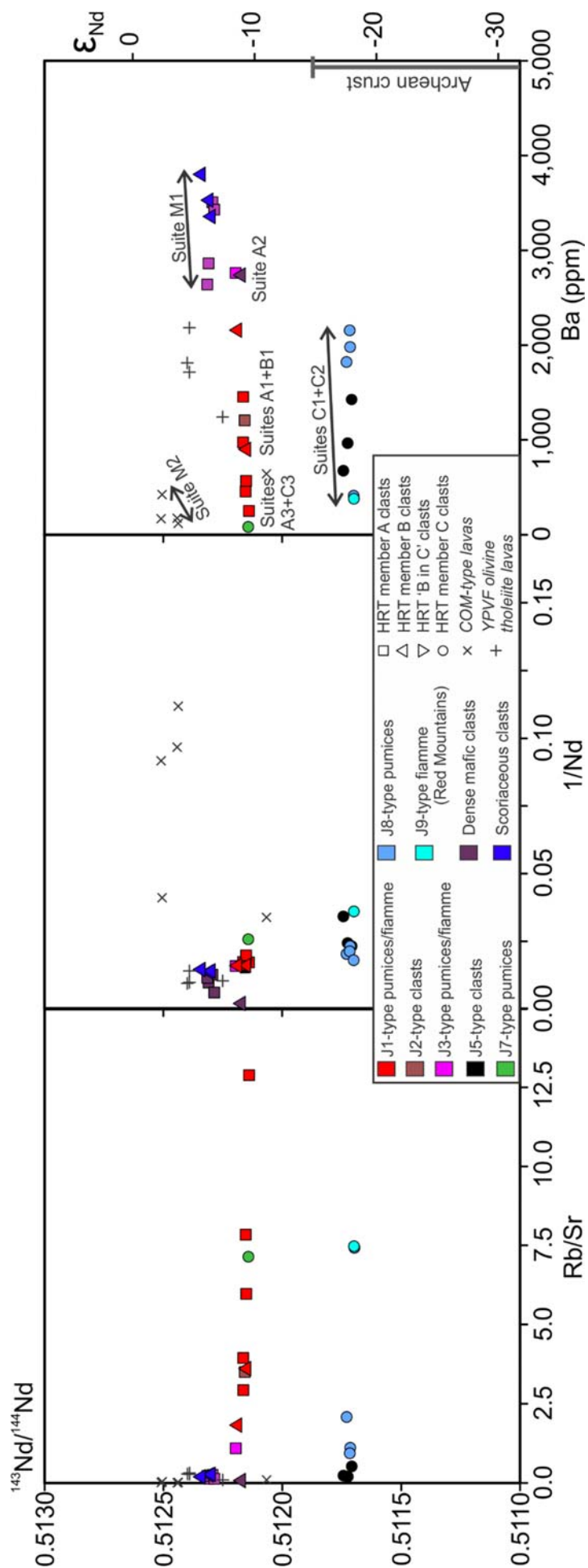


Fig. S9. $^{143}\text{Nd}/^{144}\text{Nd}$ versus indices of magmatic evolution for single clast samples from the HRT and relevant regional eruptives. Members A and B compositions lie at the un-radiogenic end of mafic suites. Isotopic compositions show no significant change with evolution (e.g. increasing Rb/Sr, decreasing Ba: cf. Sr isotopes [Fig. 14 in main paper]). Suite C3 samples are comparable to their suite A3 comparators. Suites C1 and C2 are remarkably less radiogenic but similarly show flat trends with degrees of evolution. Individual 2 se analytical uncertainties are smaller than symbol size unless shown. Other information as in Fig. 5 in the main paper. See Electronic Appendix 3 for the full data set.

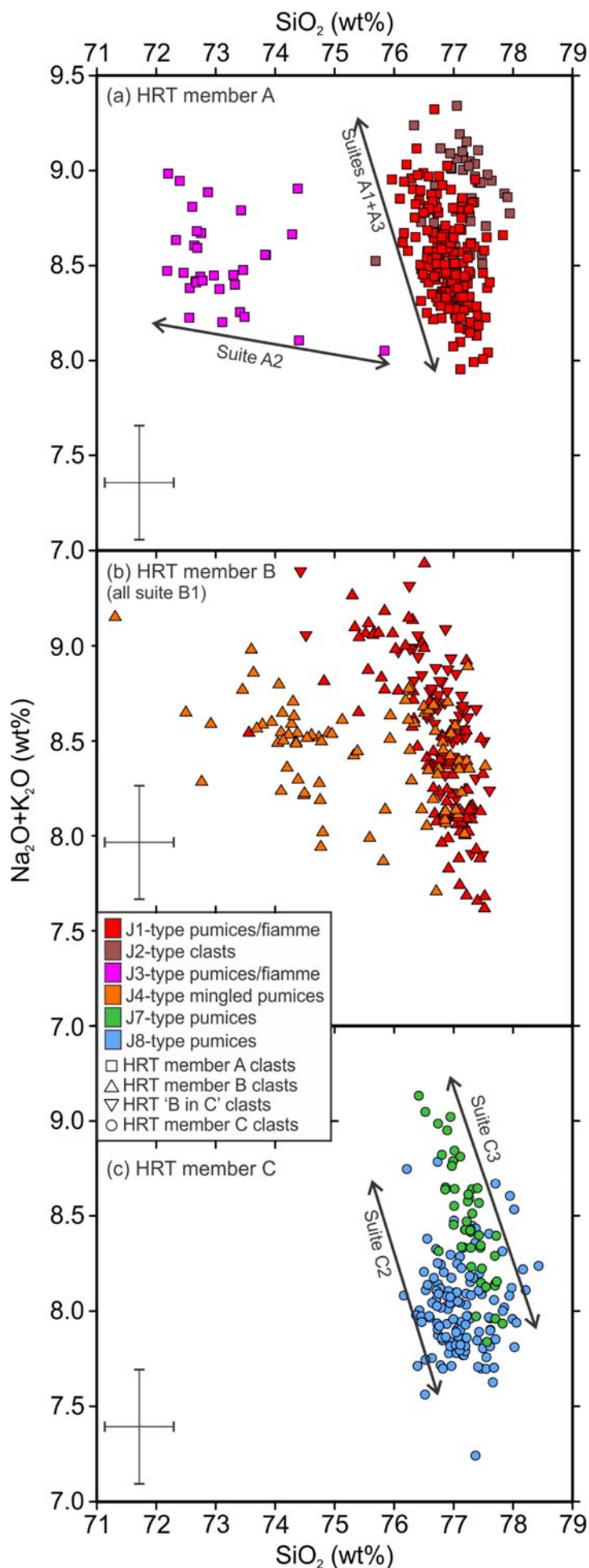


Fig. S10. Groundmass glass SiO_2 versus total alkalis ($\text{Na}_2\text{O} + \text{K}_2\text{O}$) for samples from members A, B and C of the HRT. Groundmass glass major element compositions are high-silica rhyolite, are tightly clustered and relatively homogeneous (cf. trace element compositions: Figs. 16 and 17 in main paper). Exceptions to this are type J2 samples, which may have experienced some excessive secondary hydration, and lower- SiO_2 suite A2 and type J4 scoria-bearing pumices. Cross at the bottom left of the member C plot shows 2 s.d. analytical precisions. Other information as in Fig. 5 in the main paper. See Electronic Appendix 4 for the full data set.

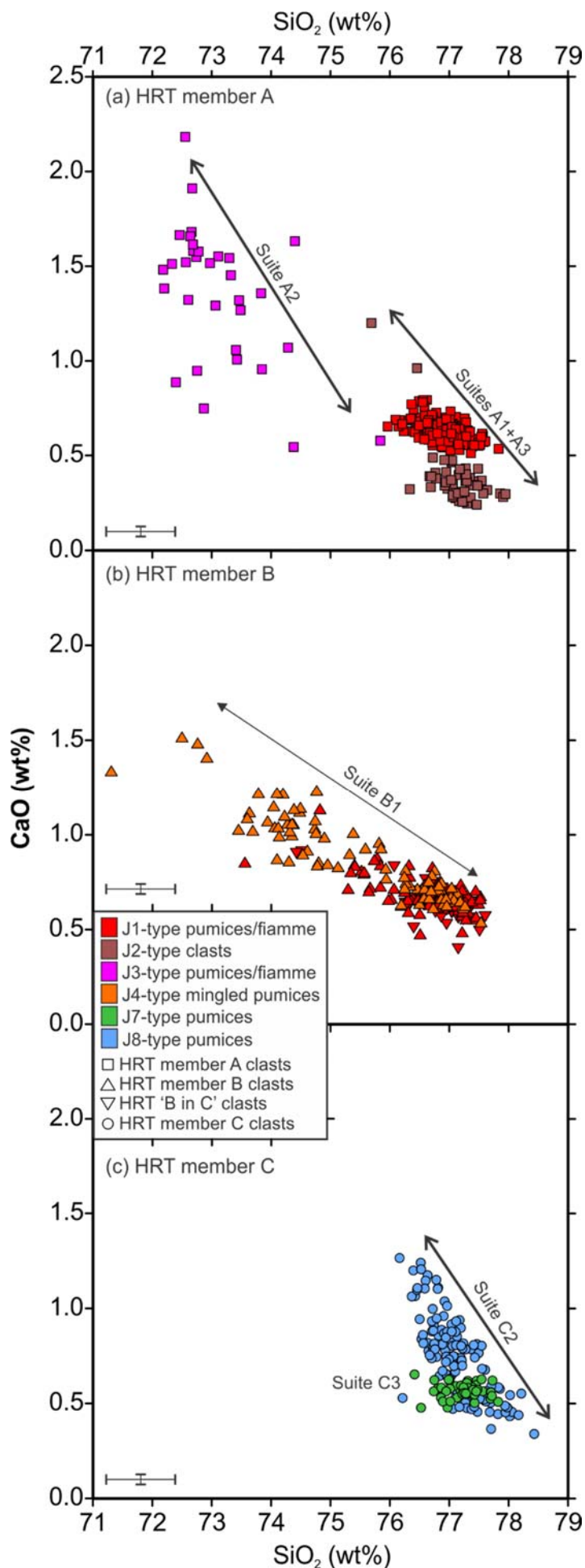


Fig. S11. Groundmass glass CaO versus SiO₂ for samples from members A, B and C of the HRT. Groundmass glass major element compositions are high-silica rhyolite, are tightly clustered and relatively homogeneous (cf. trace element compositions: Figs. 16 and 17 in main paper). Lower CaO glass from type J2 clasts may reflect excessive secondary hydration. Suite A2 and type J4 scoria-bearing pumices extend to lower-SiO₂ contents. Suite C2 groundmass glass extends to higher CaO than suite C3. Cross at the bottom left of the member C plot shows 2 s.d. analytical precisions. Other information as in Fig. 5 in the main paper. See Electronic Appendix 4 for the full data set.

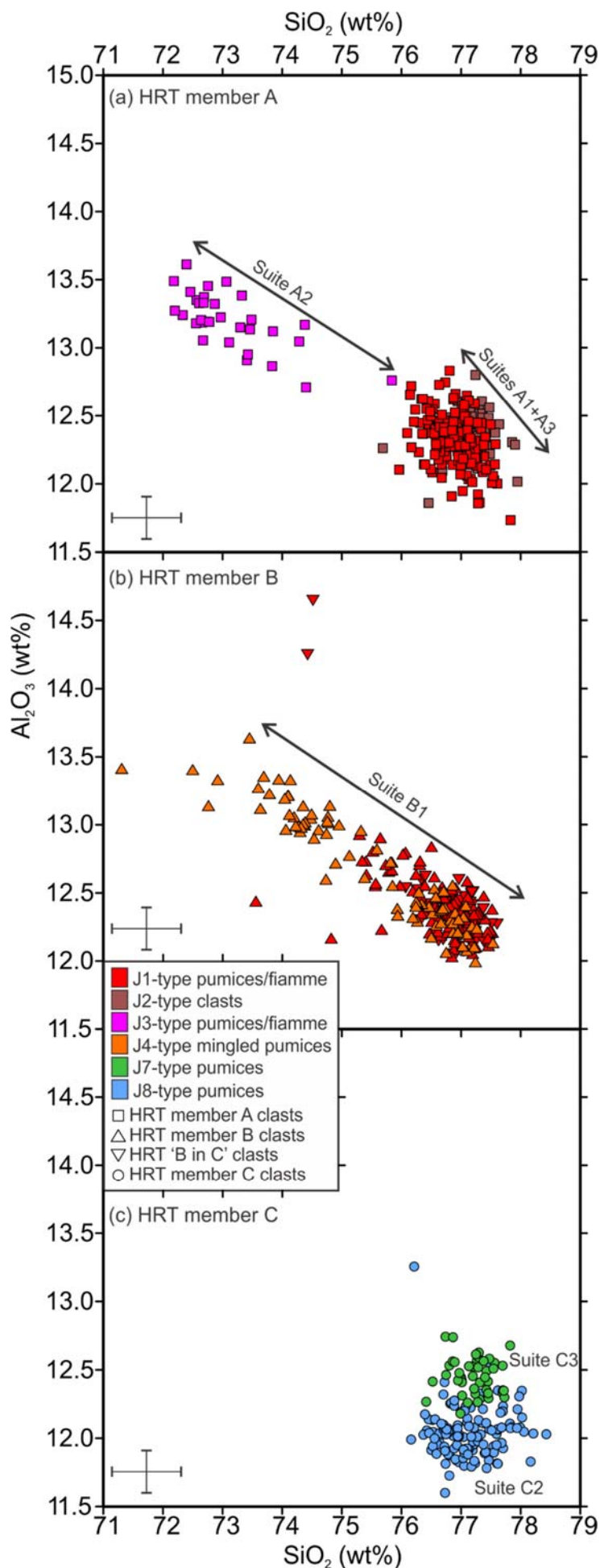


Fig. S12. Groundmass glass SiO_2 versus Al_2O_3 for samples from members A, B and C of the HRT. Groundmass glass major element compositions are high-silica rhyolite, are tightly clustered and relatively homogeneous (c.f. trace element compositions: Figs. 16 and 17 in main paper). Suite A2 and type J4 scoria-bearing pumices extend to lower- SiO_2 contents. Suite C3 groundmass glass is offset to higher Al_2O_3 than suite C2. Cross at the bottom left of the member C plot shows 2 s.d. analytical precisions. Other information as in Fig. 5 in the main paper. See Electronic Appendix 4 for the full data set.

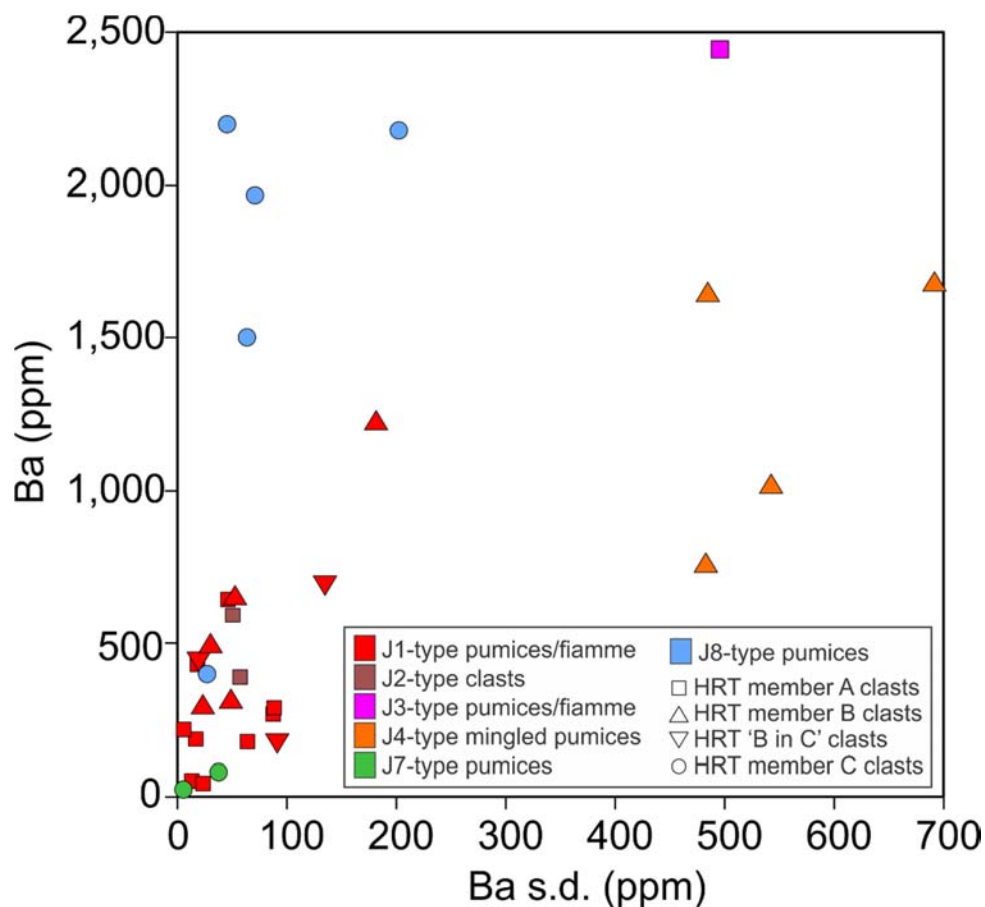


Fig. S13. Average groundmass glass Ba concentrations from a single clast versus the standard deviation (s.d.) of the variation in glass compositions within those single clasts. Groundmass glass compositions from samples in member B are typically more diverse than those from pumices in member A, particularly in the type J4 scoria bearing clasts. 2 s.d. analytical precisions are smaller than symbol size. Other information as in Fig. 5 in the main paper. See Electronic Appendix 4 for the full data set.

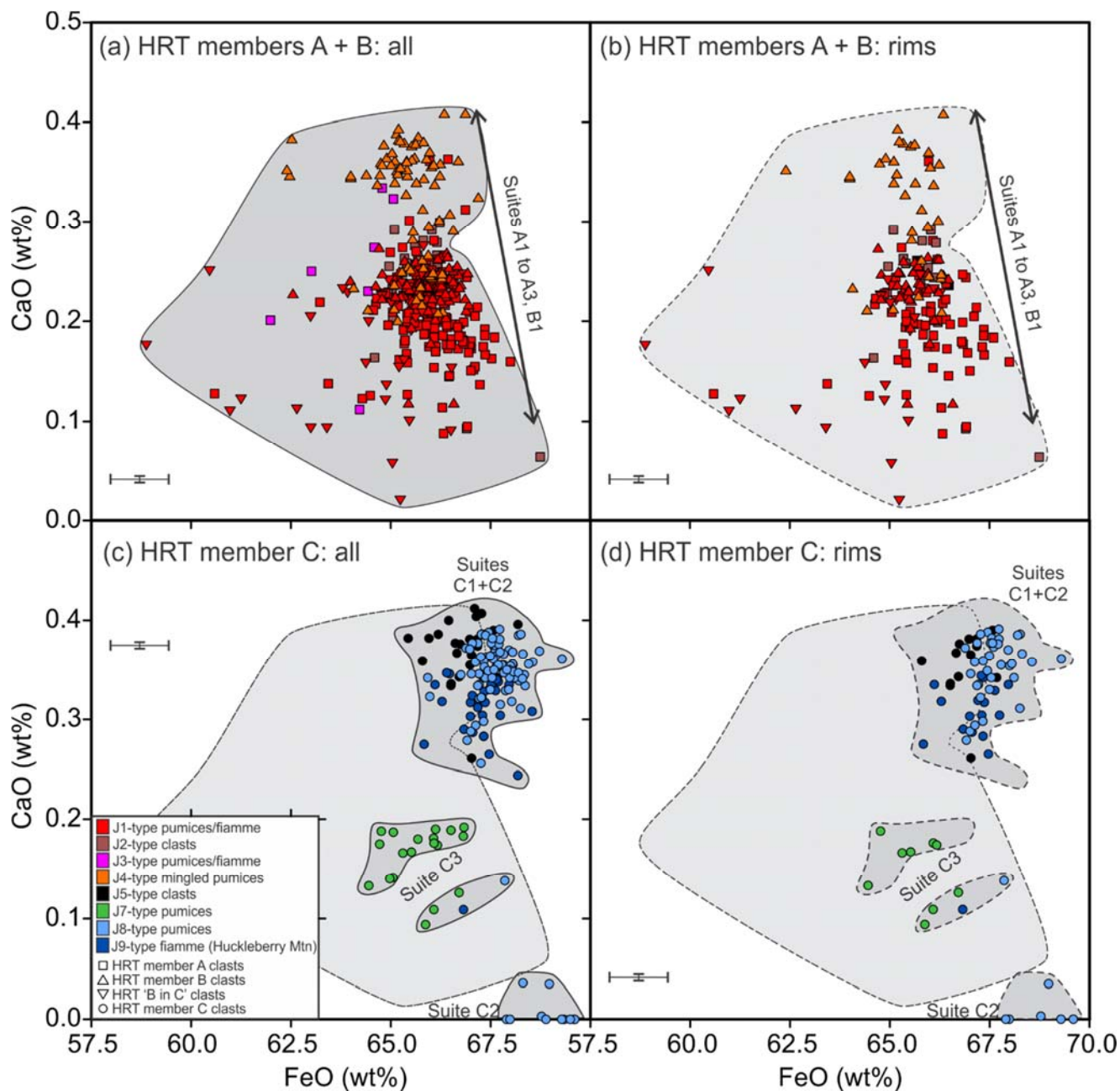


Fig. S14. CaO versus FeO for olivine compositions of crystals from the HRT. (a) All analyses from members A and B. A broad bimodality is present with a Ca-rich cluster dominated by crystals from J4-type scoria-bearing pumices. (b) Rim compositions of crystals from members A and B. Rim compositions encompass the same compositional range as interior domains. Compositional fields from (a) copied across for comparison. (c) All analyses from member C of the HRT. Suite C3 compositions cluster within the field of member A and B olivines. Suites C1 and C2 crystals are predominantly Ca-rich and offset to higher FeO members A and B. Compositional fields from (a) copied for reference. (d) Rim analyses from member C ignimbrite. A similar pattern is observed to that from all analyses. Compositional fields from (a) and (c) copied across for comparison. Cross in bottom left corner of (a) represents 2 s.d. analytical precisions. Other information as in Fig. 5 in the main paper. See Electronic Appendix 5 for the full data set.

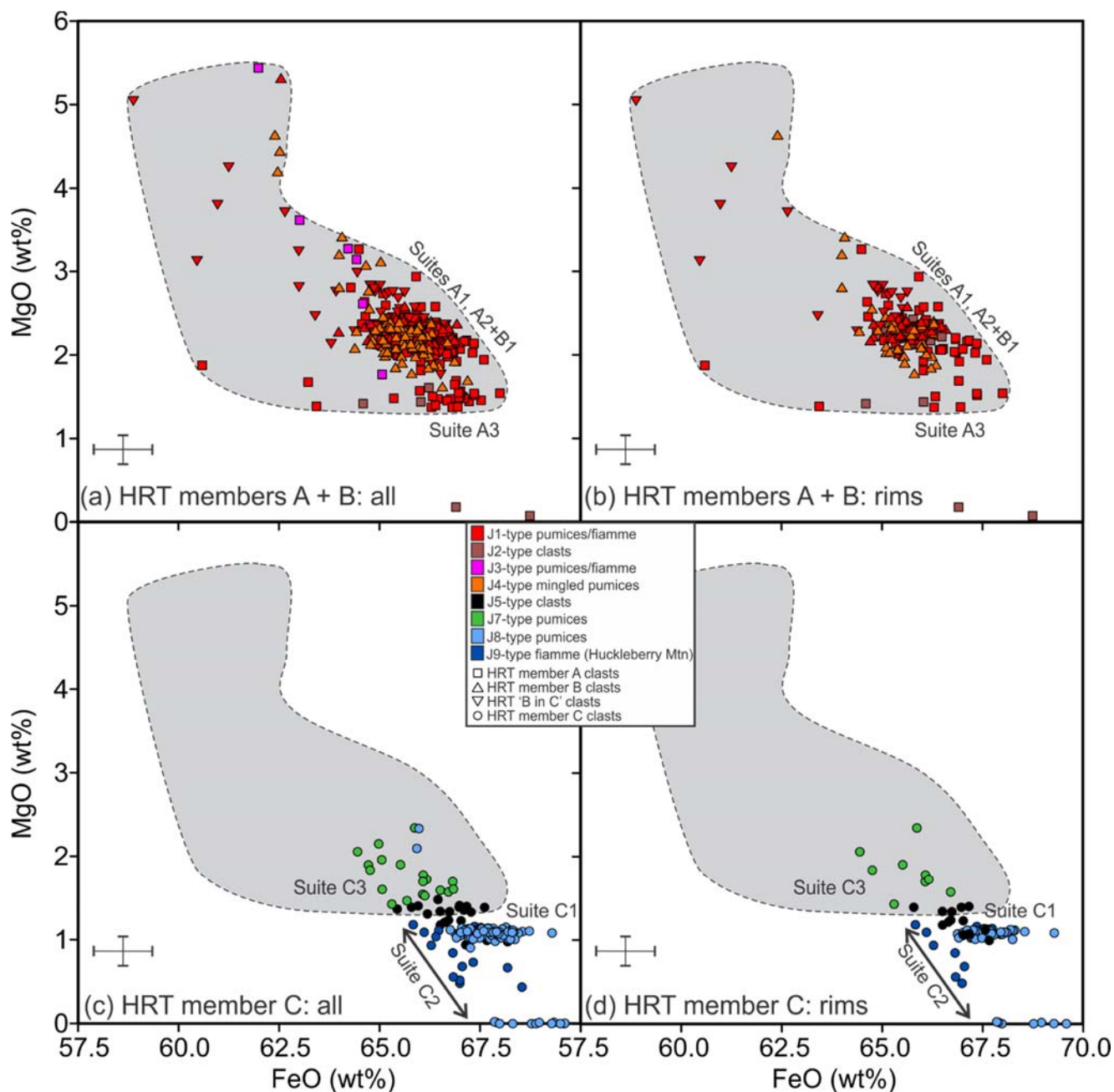


Fig. S15. MgO versus FeO for olivine compositions of crystals from the HRT. (a) All analyses from members A and B. A broad bimodality is present with a lower MgO group dominated by suite A3 crystals. (b) Rim compositions of crystals from members A and B. Rim compositions exhibit a comparable range and pattern to all analyses, consistent with the homogeneous appearance of HRT olivines. Compositional fields from (a) copied across for comparison. (c) All analyses from member C of the HRT. Clustering is evident, based on suite type. Suite C3 olivines are similar in composition to their comparable suite A3 crystals. Suites C1 and C2 crystals are offset to lower MgO with a cluster of suite C2 olivines that are MgO-free. Compositional fields from (a) copied for reference. (d) Rim analyses from member C ignimbrite. A similar pattern is observed to that from all analyses. Compositional fields from (a) and (c) copied across for comparison. Cross in bottom left corner of (a) represents 2 s.d. analytical precisions. Other information as in Fig. 5 in the main paper. See Electronic Appendix 5 for the full data set.

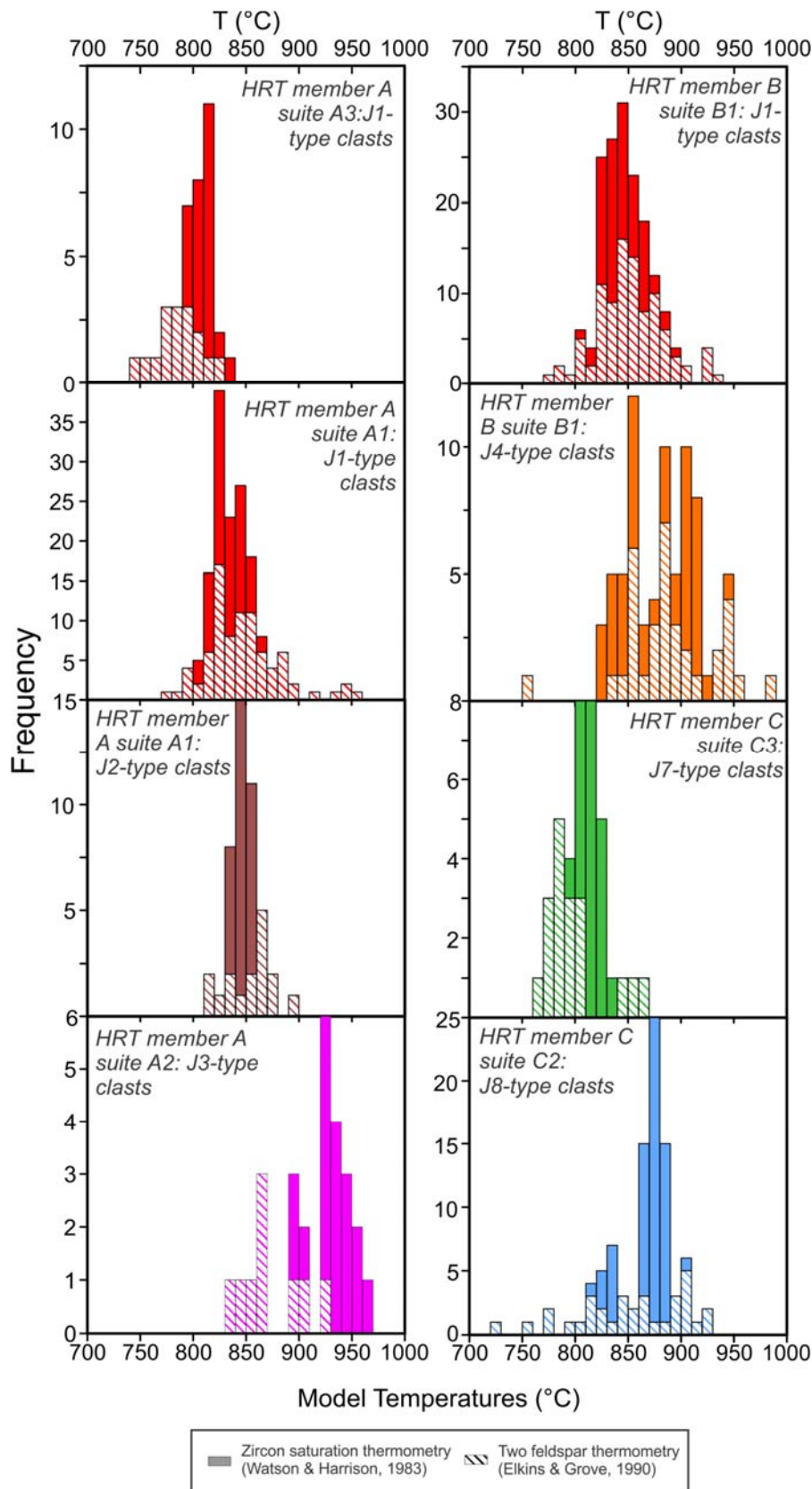


Fig. S16. Model temperature estimates from different clast types from members A, B and C of the HRT. Temperatures estimates are derived using two feldspar (Elkins & Grove, 1990) and zircon saturation (Watson & Harrison, 1983) thermometry. Suite A3 samples yield cooler temperatures than suite A1 and B1 samples, and are similar to comparable low-Ba suite C3 pumices. Zircon saturation temperatures from suite A2 clasts are offset to elevated temperatures. Type J4 mingled pumices yield a wide temperature range, encompassing suites A1, A2 and B1 (type J1 clast) temperature estimates. Suite C2 samples yield a multi-modal temperature distribution consistent with a large compositional range. Frequencies from the two datasets are stacked. See text for details, and Electronic Appendix 4 for values.

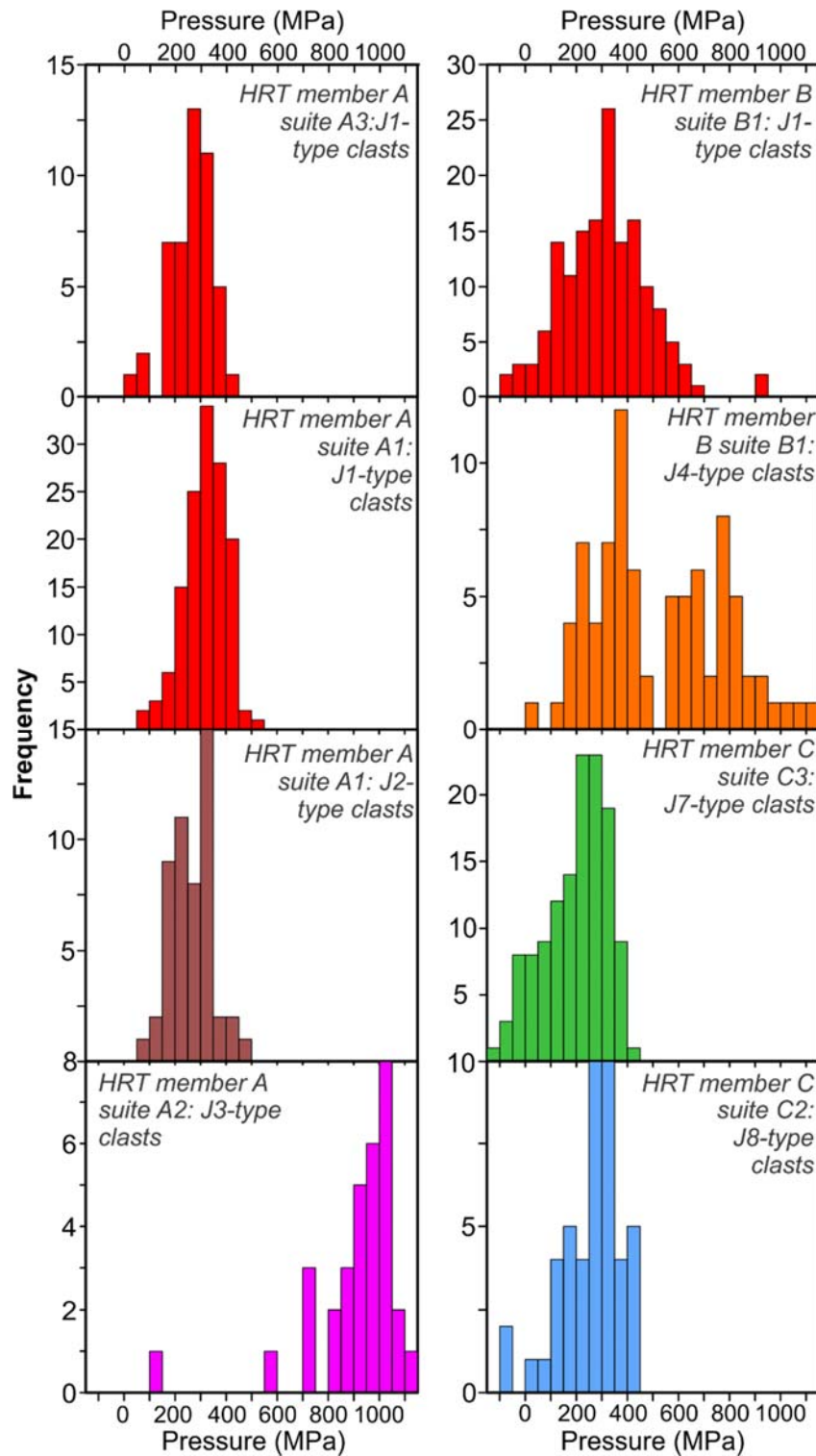


Fig. S17. Model pressure estimates from different clast types from members A, B and C of the HRT. Suites (A1, B1, etc.) are defined from compositional groupings (see text for details). Pressure estimates are derived from groundmass glass major element compositions using the calibration of Wilke *et al.* (2017; but see also Gualda *et al.*, 2019; Wilke *et al.*, 2019). Similar modal pressures are observed in suites A1, A3 and B1 (type J1 pumices). Suite A2 samples yield elevated pressures, although this is out of the range of the calibration (Wilke *et al.*, 2017). Type J4 mixed pumices yield bimodal pressure estimates, with one peak consistent with suites A1, A3 and B1, and another broader peak of elevated pressure. Suites C2 and C3 have overlapping pressure ranges, consistent with peaks observed in suites A1, A3 and B1. See text for details and Electronic Appendix 4 for values.

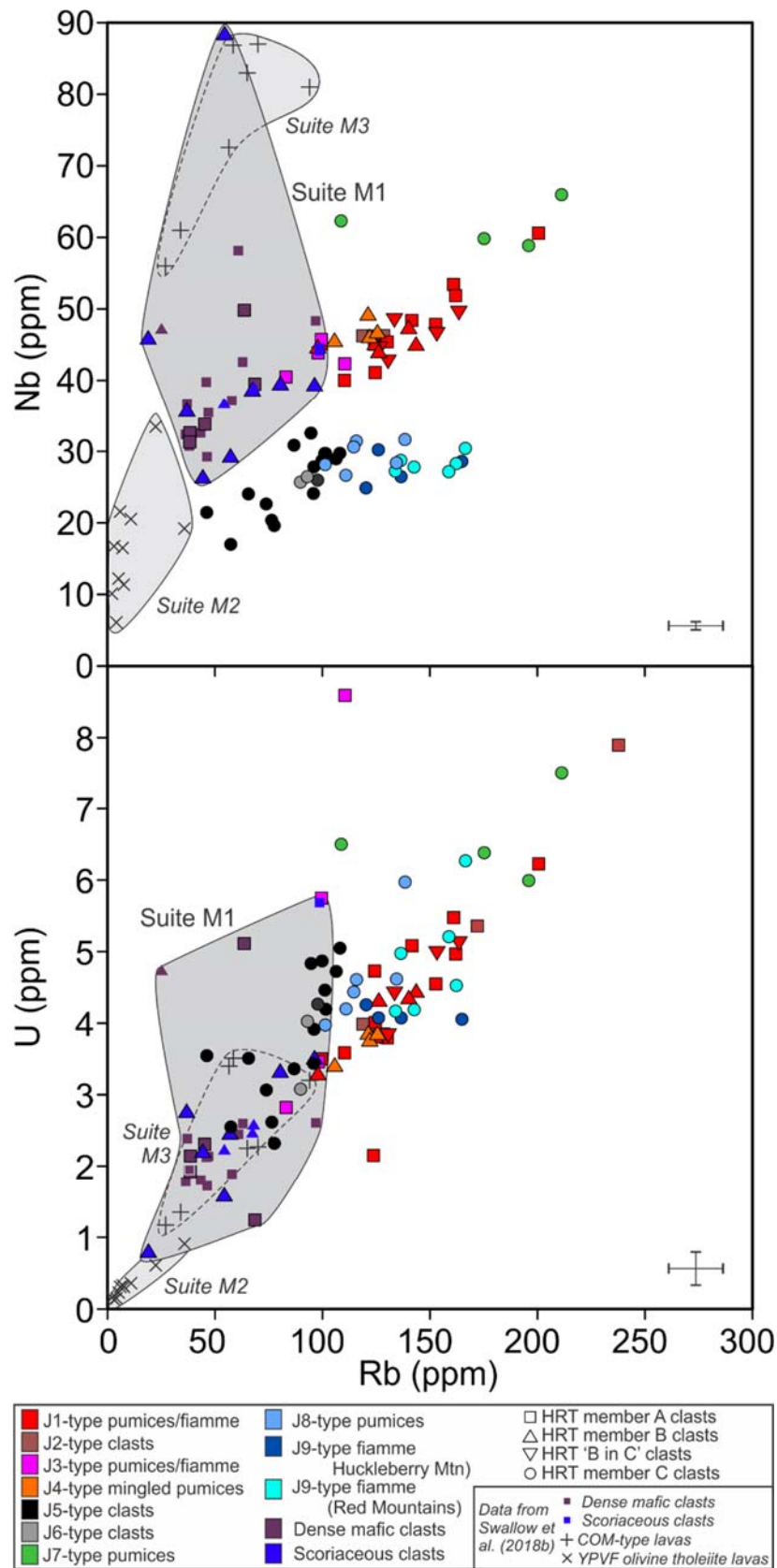


Fig. S18. Rb versus Nb (top) and U (bottom) for samples analysed in this study for the entire HRT and relevant regional mafic eruptives. Despite the major element compositional gaps (Figs. 5-7 in main paper and Figs. S2-S4), incompatible trace elements are continuous between suite M1 and suites A1-3. Crosses in the bottom right corner of the plots show analytical 2 s.d. precisions. Other information as in Fig. 5 in the main paper. See Electronic Appendix 3 for the full data set.

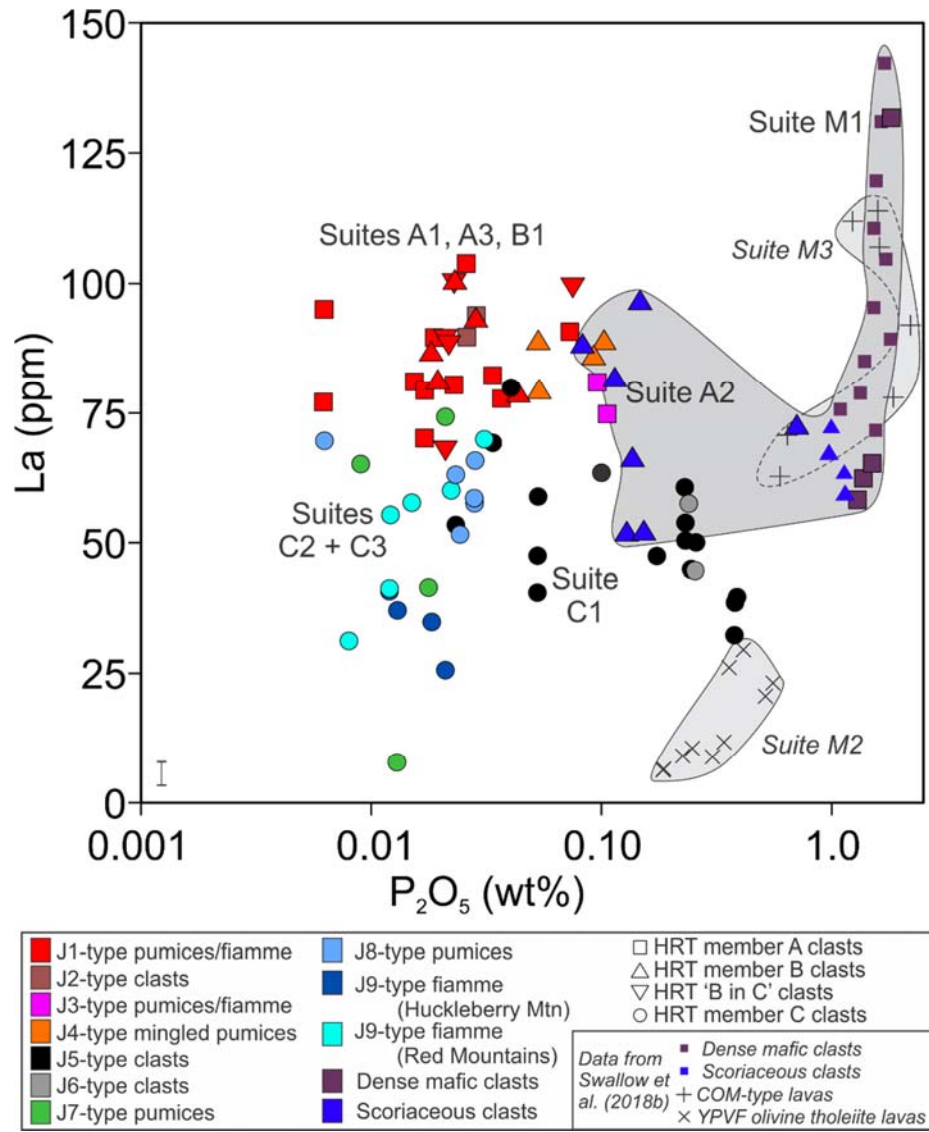


Fig. S19. La versus P_2O_5 for samples analysed in this study for the entire HRT and relevant regional eruptives (excluding anomalously high values represented in Fig. S8). P_2O_5 and La show a coupled decrease in Suite M1, indicative of apatite fractionation. Although P_2O_5 contents continue to decrease, La concentrations become decoupled and increase between suite A2 and suites A1, A3 and B1. Behaviour between the two elements is broadly coupled within suites A1, A3 and B1, possibly indicating minor chevkinite fractionation. Bar in bottom left corner indicates analytical 2 s.d. precisions for La concentrations. 2 s.d. analytical precisions for P_2O_5 measurements are smaller than symbol size. Other information as in Fig. 5 in the main paper. See Electronic Appendix 3 for the full data set.

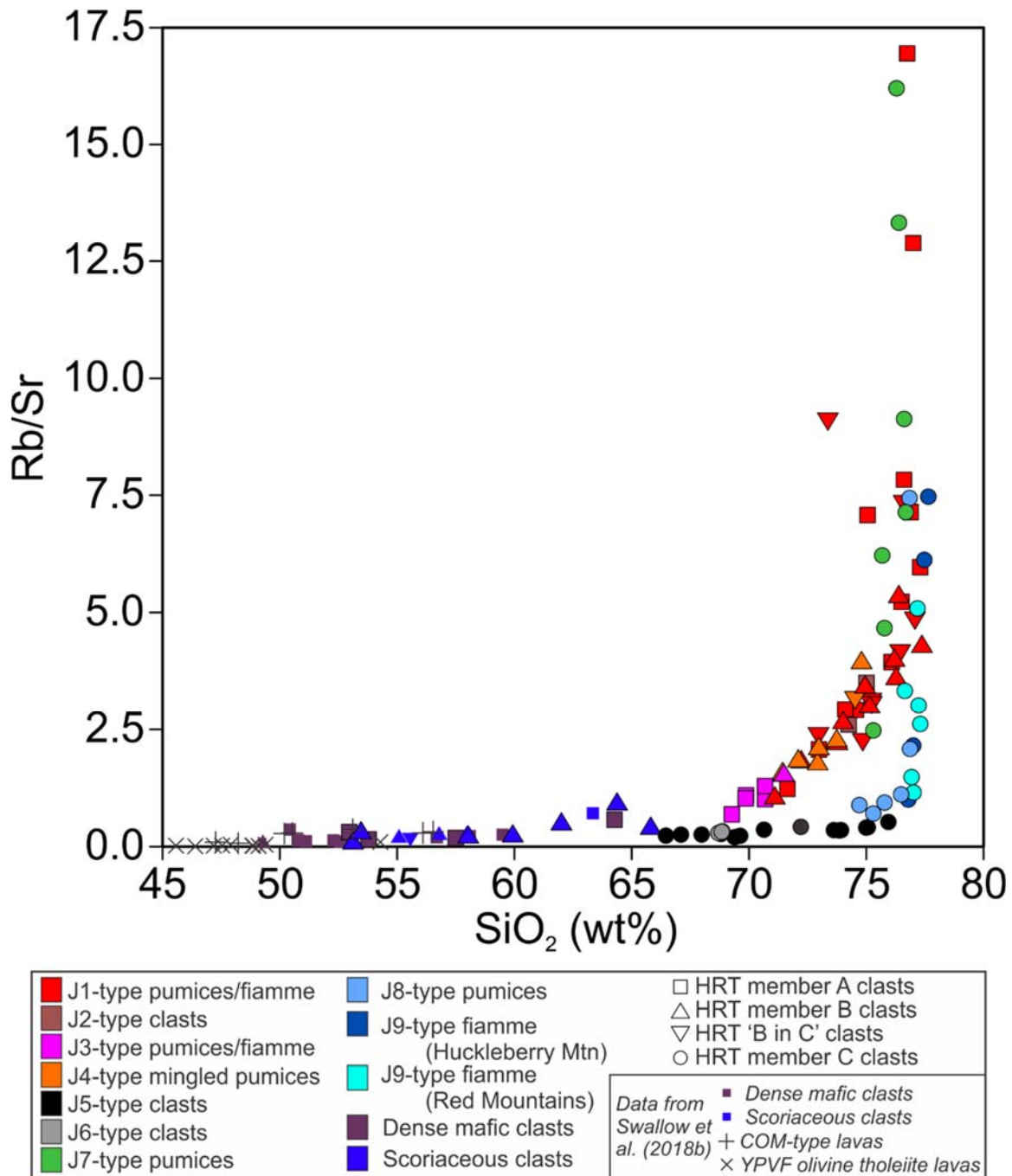


Fig. S20. SiO₂ versus Rb/Sr for samples analysed in this study for the HRT and relevant regional mafic eruptives. Of the suites that show a large compositional range, suites A1 and C2 exhibit curved trends with increasing Rb/Sr with SiO₂, indicative of fractional crystallisation. In contrast, J4-type scoria bearing-samples in suite B1 show a straight trend, typical of magma mixing. 2 s.d. analytical precisions are smaller than symbol size. Other information as in Fig. 5 in the main paper. See Electronic Appendix 3 for the full data set.

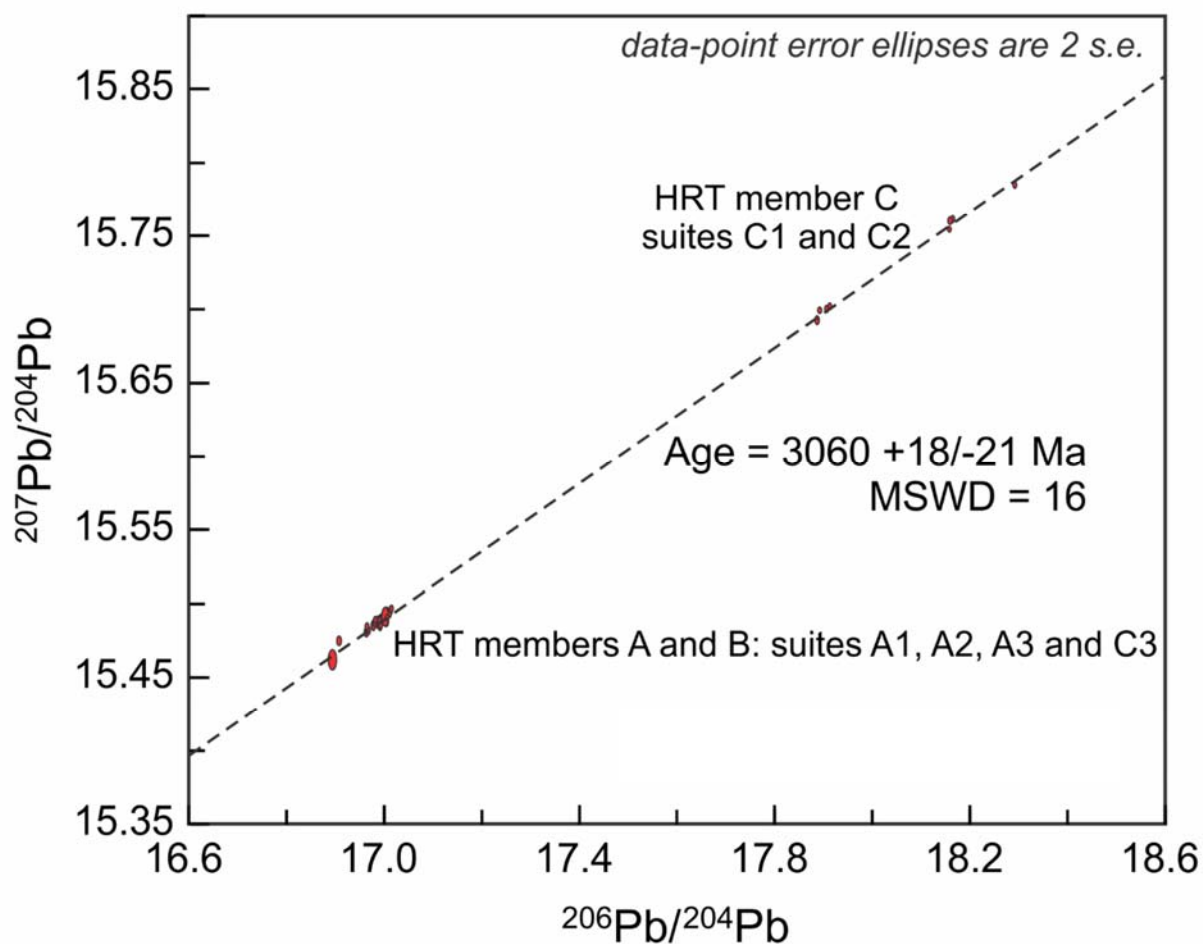


Fig. S21. Pb-Pb isochron generated by Isoplot (Ludwig, 2008) for Pb-isotopic data from silicic pumice/fiamme from all members of the HRT, yielding an apparent age of 3.06 Ga. Ellipses are 2 s.e. See Electronic Appendix 3 for the data set.

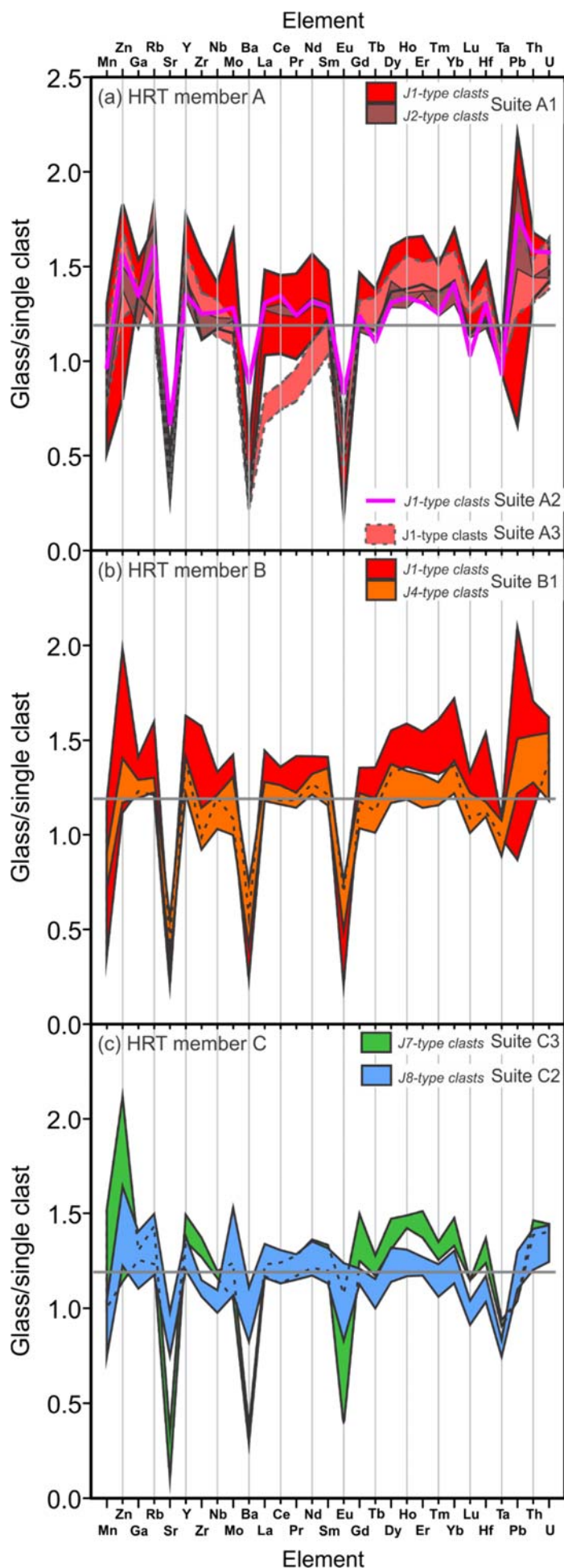


Fig. S22. Multi-element diagrams for glassy pumices from each member of the HRT showing the ratio between the average groundmass glass concentration and the whole-clast composition. The glass compositions are generally enriched (glass/whole-clast > 1) in incompatible elements. Groundmass glass from suite A3 samples is depleted in LREE (glass/whole-clast < 1) indicating minor chevkinite fractionation. In contrast, comparable compositional suite C3 pumices have LREE-enriched groundmass glass (glass/whole-clast > 1) precluding significant chevkinite fractionation. Full data are in Electronic Appendices 3 (whole-clast) and 4 (glass).

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